# University of Cincinnati

**Date: 9/1/2015**

**I, Sayantika Mukherjee , hereby submit this original work as part of the requirements for the degree of Doctor of Philosophy in Electrical Engineering.**

It is entitled:

**Fundamentals and Applications of Large Area Multi-Spectral State Electrophoretic Panels for Displays and Smart Windows**

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# Fundamentals and Applications of Large Area Multi-spectral state Electrophoretic Panels for Displays and Smart Windows

A dissertation thesis submitted to the

Graduate Faculty of the University of Cincinnati

In partial fulfillment of the requirements for the degree of

**DOCTOR OF PHILOSOPHY (PhD)**

In the Department of Electrical Engineering and Computing Systems

Of the College of Engineering and Applied Science

2015

By

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## *Abstract*

Large area electro-optic switches are increasingly important, including flat panel displays and the more recently emerging technology of smart windows. However, these types of switches are typically limited to switching between only two states, such as a clear and an opaque state. Therefore optical performance and functionality of large-area electro-optic devices could be significantly improved, if switches were developed that could provide modulation of multiple spectral states. Simple solutions such as adding a color filter array onto a clear/opaque switch are not the ideal solution, as seen in displays where color filters reduce the display brightness by blocking more than 2/3rds of the visible spectrum. There is a clear need to develop alternate electro-optic switches, especially those that broaden the spectral capability within the switch without a significant increase in complexity of the device or increase in optical loss. If such a novel switch could be realized, full-color displays could be bright enough to even be easily read in sunlight, and smart windows could be made into more compelling products by allowing additional desirable lighting features such as control of color temperature.

In this dissertation a completely new bi-primary pixel switching mechanism is demonstrated that can switch one single pixel into 4 color states (2 color states along with black and white). The approach uses two-particle, two-colored electrophoretic inks along with novel device architectures which could simplify large area device manufacturing. For displays, this approach provides 2X higher brightness and reflectivity than the already existing side-by-side RGBW color system approach. For smart windows, dual spectrum control is enabled (color temperature, visible vs. IR, etc.), finally enabling smart window technology which provides features not easily possible with simple mechanical blinds. Importantly, this new structure is highly manufacturable, requiring no alignment, and can be fabricated by micro replication and roller printing methods. This dissertation spans fundamental theory, through experimental electrophoretic characterization, through compelling integration and performance demonstrations.

## Copyright Page

## Acknowledgement

This dissertation work would not be possible without the sincere help of a few people. Firstly I would like to thank my adviser Dr. Jason C. Heikenfeld, who has been my mentor and guide and led me to successfully reach where I am today. He believed in me and always inspired me to push my limits. This dissertation work would not have been possible without his expert guidance and directions. I would also like to thank my former lab mates Dr. Matt Hagedon, who made me familiar with all the lab processes and equipments and providing me with all the basic training I ever required to perform all my future experiments, Dr. Brad Cumby, Dr. Phillip Schultz, Dr. Alex Schultz, who never showed any less enthusiasm when I ran to discuss any issues/ questions I had. They always enlightened me with logic and possible solutions to my problems. I am thankful to have worked with such an enthusiastic and bright group of people. I would also like to thank all my current lab mates Daniel Rose, Sarah Holcomb, Dr. Selva Kumar Sagaya, Aaron Diebold, Phillip Simmers, and Julian Morand who also provided me with valuable knowledge and scientific discussions.

I would like to thank our industry partners Merck Chemicals Ltd. UK, for providing materials that we required for our experiments, and Hewlett Packard Research Center USA for providing valuable guidance to reach my ultimate dissertation goal.

I would like to thank my PhD dissertation Committee Dr. Andrew J. Steckl, Dr. Peter B. Kosel, Dr. Ian Papautsky and Dr. Rupak K. Banerjee for providing me valuable suggestions and guidance on this dissertation work.

I would finally wish to extend my gratefulness to my Mom and Dad who believed in me and let me walk on this path, my sister who has been my backbone and inspiration and my friends who showed enough confidence in me that I could accomplish this task today.

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# *Chapter 1: Introduction*

## 1.1 Introduction to this Chapter

This chapter introduces the background and motivation for the dissertation. First, we evaluate the contemporary electro-optic switching techniques available and we discuss the shortcomings and advantages of those. We then introduce the concept of alternate electro-optic switching of pixels and the physical phenomenon which will enable the operation of such a technique. The goal of this dissertation is twofold: 1) implementing complementary-colored dual particle ink dispersions in an electrophoretic device structure, that will be able to provide 2X brighter color than the standard RGBW color system for display applications; (2) implementing complementary-colored dual particle ink dispersions in a similar structure, which will broaden the capability to include color tuning control for smart window applications. The device structure is aimed to be created with very few fabrication steps, to be highly scalable and micro replicable, which will reduce the overall fabrication cost.

## 1.2 Electro-optic switching

Large area electro-optic switches are used in both displays and smart window. Displays including today's most prolific ones like the Liquid Crystal Displays (LCDs) that are used in large and small sized devices utilize a backlight and a pair of cross-polarizer's to alternate between clear and a dark states per pixel [1]. A smart window alters the incoming solar spectrum between completely opaque and clear [3], [4], [5]. A review is presented of some of the contemporary technologies that utilize electro-optic switching of pixels and the drawbacks which exist. The broad categories of electro-optic switching under which the contemporary technologies will be reviewed are i) transmissive mode and ii) reflective mode.

### 1.2.1 Transmissive

Transmissive displays are those that utilize a backlight or a light source to transmit light through the device assembly, the most common example of such a display application is the liquid crystal displays or the LCDs. Smart window technology is also obviously transmissive, where the light source is outside light for external windows, or the light is in another room or area of a building for internal windows.

#### 1.2.1.1 Liquid crystal (mainly transmissive, very limited reflective use)

Liquid Crystal Displays (LCDs) are used in large and small sized devices most commonly for information displays commercially, and utilize a backlight and a pair of cross-polarizer's to switch between clear and dark pixel states.

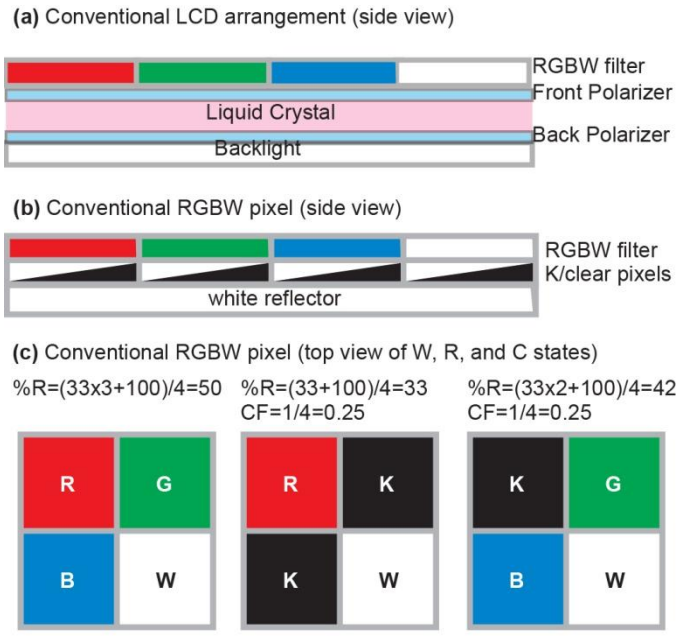


**Figure 1.1**: (a) A typical LCD arrangement (side view) (b) Conventional RGBW arrangement (c) Conventional RGBW producing W, R and C colors (top view)

The color rendering of an LCD is obtained by placing a color filter array of side-by-side Red/Green/Blue (RGB) or Red/Green/Blue/White (RGBW) pixels on the front polarizer [1]. The polarizers themselves reduce the incoming light brightness and the color filter in turn reduces the brightness even further.

The overall reflection for each color is only 33% as each RGB filter comprises 1/3 of the visible spectrum. The overall transmission or reflection only comes up to 50% theoretically with use of the white sub pixel and is worse for the RGB only pixel. This is a drawback in Liquid Crystal Displays when switching its pixels to a bright color state, and hence it leads to high power usage, poor readability under high luminance condition such as outdoor sunlight. This is also true for display technologies like Organic Light Emitting Diodes (OLEDs) that need internal built-in lights to generate images of reasonable quality. The need for internal built-in light is hence not always the best solution for illuminating pixels for switching and also these technologies described only switch pixels to either a single-color or black state.

#### 1.2.1.2 Electrochromic (mainly transmissive, very limited reflective use)

Electrochromism [2], [5] is more than 30 years old technology, and has been widely used for displays as well as transmissive smart window and reflective rear view automobile mirrors. Electrochromism is listed under transmissive type due to their use in smart window technology, but can be considered to be a reflective technology when used for displays used with a back reflector.

They have a wide variety of chemistry and performance but most of their commercial approaches have been just monochromatic. However, a full color approach can be obtained by stacking subtractive CMY layer one over the other illustrated in Figure 1.2, for which each layer switches between a color state and a clear state.

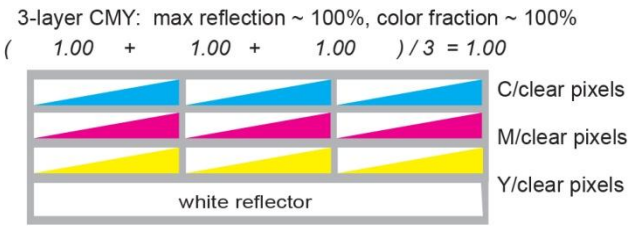


**Figure 1.2**: Stacking of subtractive CMY layer for a typical electrochromic display switching mechanism

Theoretically each layer now provides 100% reflection but the complexity arises due to multilayer stacking that gives rise to complex fabrication steps, requirement of alignment of thin flexible substrates, complex driver electronics, parallax error and contrast reducing glare due to stacking of transparent substrates. These issues can cause optical loss to become enormous at high resolution and result in poor resolution and low brightness images.

#### 1.2.1.3 Suspended particle device (transmissive, reflective possible but not used)

Suspended particle device or light valves are currently used in smart windows for transmitting or blocking incoming light spectrum [6]. The suspended particles are suspended in a liquid film where they can move freely, in between two panels of glass or plastic coated with transparent conductive material as illustrated in Figure 1.3. They remain randomly arranged, therefore blocking any light spectrum to pass through without any applied voltage. But when a voltage is applied, the particles align themselves such that the light spectrum passes through them making the panel transmissive. When the amount of voltage is decreased the particles spread randomly again and the window starts to darken until the light is completely blocked. This switching technique also only switches between clear and opaque. This is a restricted functionality of the current state of smart windows as windows are essentially a source of lighting. Unfortunately, switching between only a clear or opaque state has little performance advantage over traditional mechanical blinds. Such limitation sets up part of the motivation for pursuing the work of this

dissertation. For example, attractive capability could include color-temperature control of the incoming solar spectrum along with completely transmissive and complete opaque states, for which clearly a multi-spectral switching device is needed.

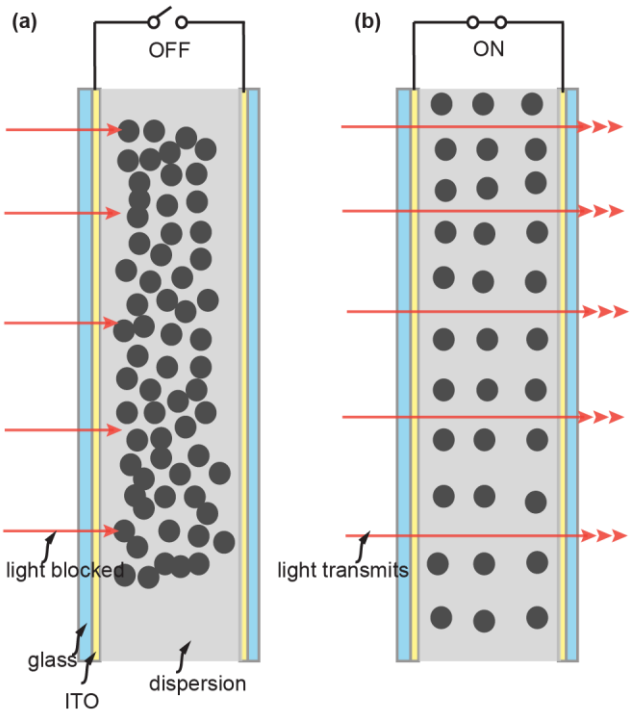


**Figure 1.3**: A typical suspended particle device switching of smart window (a) opaque (b) clear

### 1.2.2 Reflective

Reflective type displays are those that do not require a backlight or a source of light for illuminating the pixels. They instead utilize a paper like reflector at the back to scatter the ambient light. The Amazon™ Kindle e-reader is one such example. Several of the examples listed below can also be optically transmissive, but are listed here because their main use is reflective operation.

#### 1.2.2.1 Electrowetting (reflective mainly, transmissive possible)

Electrowetting technology was first reported by Philips research labs for reflective displays [1]. The operation includes the change in area coverage of colored oil caused by wetting and de-wetting of the device surface by a transparent conductive (polar) medium (Figure 1.4)

The dyed oil is transposed between a hydrophobic dielectric film (no voltage) and a partial sphere reduced to 20-30% of the pixel area (voltage). The switching thus consists of wetting and dewetting oil droplet hence that can still be done monochromatically and a full color operation is possible by placing a color filter array over the monochromatic pixels. Electrowetting has been implemented for transmissive smart window switching as well but is still limited to opaque to clear switching states only [4].

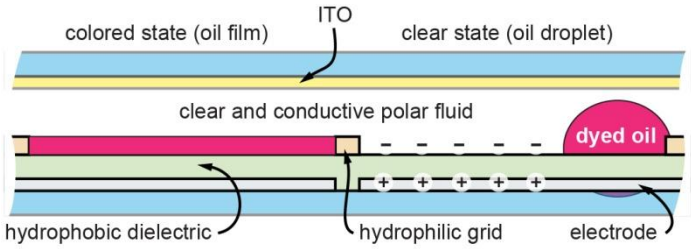


**Figure 1.4**: Illustration of a typical Electrowetting pixel switching between clear and color (reproduced from [1]). A reflector is placed beneath the device to enable reflective mode or the bottom electrode is constructed of a reflective metal such as aluminum.

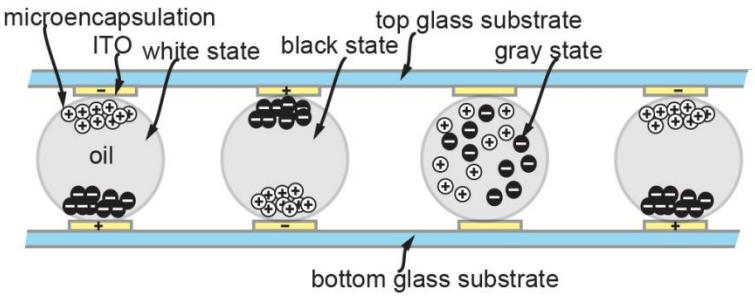


**Figure 1.5**: A typical microencapsulated vertical electrophoretic b/w particle switching that E-Ink uses

#### 1.2.2.2 Electrophoretic mainly E-Ink (reflective only)

The reflective displays or Electronic paper solves the problem by offering low power and high readability in outdoor lighting conditions [1],[7]. They do not employ any internal light source and rely on ambient light only. The current dominating technology in the fields of electronic paper (e-paper) is the Kindle™ E-reader that uses E-ink. E-Ink technology switches pixels by electrophoretic transposition of black and white pigment particles that are microencapsulated (Figure 1.5). It has become highly popular due to its high reflectivity and the appearance of a real print on paper. But the switching of the pixels is merely black or white and a full color generation is only possible by placing a RGBW color filter array on top of the black and white pixels similar to LCDs, which again results in very poor reflectivity and low brightness.

Clearly an alternate electro-optic switching technique for the pixels are required that can switch the pixels into multi color state without the loss in reflectivity or brightness. Again, this need sets up the motivation for this dissertation.

### 1.2. 3 The Concept of Colorant Transposition for Electro-optic Switching

As described above for electro-optic switching of pixels, existing technologies simply switch monochromatically (clear or color/opaque) which limits the performance and functionality

depending on the applications. The technology used in E-Ink is electrophoresis which transposes particles through a fluid in a uniform electric field. If only this technology can be employed with multiple types of colored particles instead of just black/white, pixels can be switched to bright color states without the compromises made in brightness, reflectivity or functionality. Such a technique is described in the figure 1.6. By employing dual color dispersions where we can move around the two colored particles such that when they are completely mixed gives a mixed state, when both of them can be hidden in a small area of the pixel in two sides can give us a clear pixel, and by hiding one color and spreading the other one can give us two more different color states.

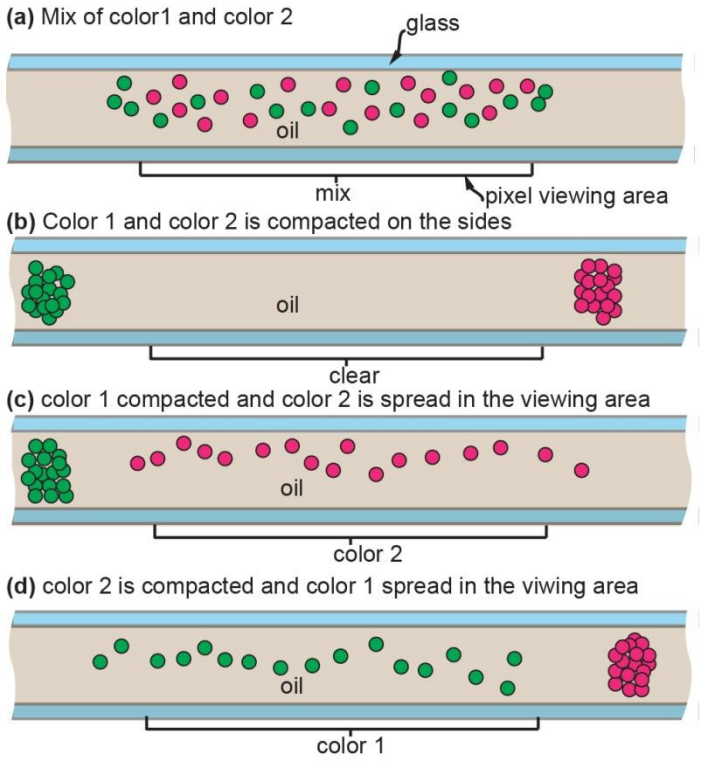


**Figure 1.6**: A technique by which 4 types of switching can be obtained (a) mix (b) clear (c) color2 and (d) color1

For such a colorant transposition containing two particles can be done using electrophoresis which is discussed in detail in the next section. The concept of switching two different colored particles is not new, and has been demonstrated in in-plane only devices by Philips research labs [ref]. However, the switching performance of such devices is extremely challenged in both switching speed and in fabrication complexity (and cost) for low-cost / large-area applications. This dissertation will provide results which go well beyond the start-of-the art for two-colored particle switching, including the 1st device designs which are proposed to be highly manufacturable over large areas.

### 1.3 Electrophoresis Fundamentals

Electrophoresis involves the motion of charged colloidal particles relative to the dispersion fluid in which they are dispersed, due to application of a uniform electric field. Electrophoresis is an essential technology in microfluidics and is essentially used for separation of macromolecules based on size e.g. DNA and RNA analysis. Electrophoresis of positively charged particles is called cataphoresis and of negatively charged particle is called anaphoresis. The application of electrophoresis in displays is also discussed in detail in the following sections.

#### 1.3.1 Electrophoresis Theory

In an electric field, a charged particle in a fluid moves in response to an applied E field with a velocity v relative to the surrounding fluid. The proportionality constant μ is called the electrophoretic mobility.

$$v = \mu E$$

The charged particles can essentially be charged molecules or charged colloidal particles. Figure 1.7 show electrophoresis on spherical charged particles with charge q in an uncharged viscous medium. The resultant velocity of the particle is dependent on the balance between the electrostatic force ($F_E$) and the frictional force ($F_{FRICTION}$). For a particle in an electrolyte, the net

velocity with which it moves depends on the balance between the electrostatic force, the frictional force and forces caused due to retardation and relaxation of the countercharges present due to the electrical double layer around the particle in the electrolyte.

Smoluchowski first developed an expression for the electrophoretic mobility

$$\mu = \frac{\zeta\varepsilon}{\eta}$$

Where **ε** and **η** are dielectric constant and viscosity of the surrounding fluid, respectively, and **ζ**, the so called zeta potential is also the electrostatic potential at the shear plane around the particle. This assumption was for a very thin double layer around the particle. But for very low dielectric dispersion medium and a thicker double layer Huckel developed the expression:

$$\mu = \frac{2}{3} \times \frac{\zeta\varepsilon}{\eta} \times F(Ka)$$

Where $K^{-1}$ is the Debye length and a is the radius of the particle, F is a function that is approximated to 1 for simplicity and for particle size <1μm

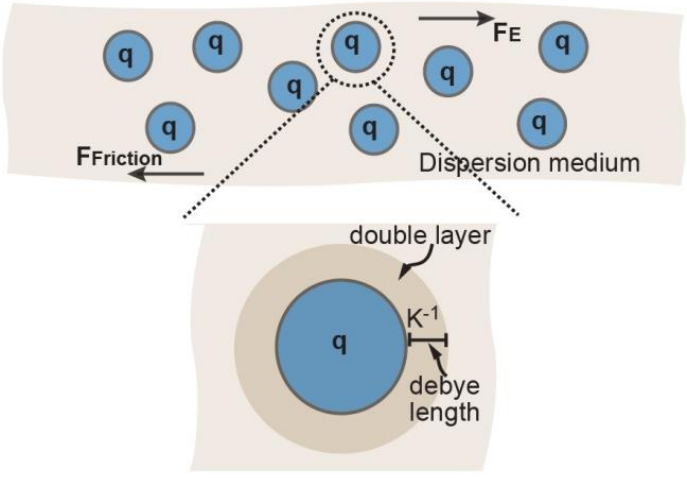


**Figure 1.7**: Charged colloidal particles in a dispersion medium for electrophoretic movement

The particle movement is driven by an impulse (field*time), so that to the first order the response time of a pixel is given by

$$t = \frac{d^{2}}{\mu V}$$

Where d is the cell gap and *V* the voltage applied. These equations show how particle and fluid properties are important for fast switching. Also the voltage applied and the cell gaps are other important parameters to consider along with the ones mentioned for a reasonable switching time period.

### 1.3.2 Electrophoretic Switching of pixels

Optical switching with electrophoretic particles is currently being explored mostly in the field of the reflective displays e.g.; the Amazon™ Kindle e-reader that uses E-Ink. Let us now review in more detail than found in previous sections, the technologies available in the field of electrophoretic switching of pixels.

#### 1.3.2.1 Vertical electrophoretic switching

Microencapsulated electrophoretic display films been developed by E-Ink Corporation [7] and independently at the NOK Corporation, and uses the phenomenon of vertically transposing electrophoretic particles towards or away from the viewing area. E-Ink's first microencapsulated electrophoretic film had a particle-dye formulation. The bright state was achieved by white scattering particles and the dark state was achieved with a dye in the electrophoretic fluid. The recent electrophoretic films made at the E-Ink Corporation have a dual-particle formulation where the dark colored state is achieved by a second particle that is black in color. Apart from black/white, subsequent gray levels can be achieved by partial addressing between the black and white states. This system is much better in color contrast as per b/w monochrome e-reader applications, and is used in the manufacture of Amazon-Kindle™ (see Figure 1.5). Due to its

low power operation and print on paper like appearance, the requirement of a colored prototype is still being researched. Color display prototypes using E-Ink's electrophoretic films are being made in conjunction with a color filter and was demonstrated [8], [9]. But as already discussed, this color-filter approach does not provide high brightness or contrast. So it is desirable, to use real color pigments instead of color filters to provide the high contrast and brightness required to mimic the print on paper mode. Although, in principle it should be possible to use combinations of colored pigments to generate full color, these approaches have not yet been successful [1]. The electrophoretic display (EPD) currently employing E-Ink technology has achieved a reasonable maturity with a white state of 40% and a contrast ratio of 10:1. The vertical EPDs are bistable, and in that they maintain a grayscale image for a long period of time after removing the signal (days or more). The power consumption is also quite small because the images are on updated as needed which is often only every minute or so [10].

#### 1.3.2.2 In-plane electrophoretic switching

The horizontal or in-plane electrophoretic display operates through the lateral movement of the pigments across the pixel, first reported by IBM and Canon [11]. The colorant particles absorb light when distributed across the pixel giving a colored state, and a clear state when the particles are collected onto a small area which is hidden from the viewing area. Canon reported an active matrix in-plane electrophoretic display of 200 PPI in 2002 [12]. Philips reported pixelated displays without an active matrix using one or two charged colorants through the use of a gate electrode. With the ability to achieve a clear or light transparent state, multiple layers can be stacked with subtractive CMY colorants to achieve bright color electronic paper [13]. But the subtractive colorant approach results into unreasonable optical loss and parallax error, and the net color output is also poor. Unlike vertical switching, in-plane switching has not yet shown bistability, but the holding of the pigments in in-plane switching typically requires a very low

voltage and hence they are suitable for low power operations, Figure 1.8 (a) illustrate the in-plane switching phenomenon.

#### 1.3.2.3 Electrokinetic switching

HP developed a more sophisticated architecture of electrophoretic pixel switching and demonstrated them first time at the SID 2004 international symposium on display technology. This is a hybrid version between the in-plane and vertical electrophoretic switching, as illustrated in Figure 1.8 (b). Micro-pits allow compaction and isolation of pigment particles inside the pixels [14] [15]. The pigments move both vertically and also in-plane to the micropits. The advantages of this kind of architecture includes fast switching, better compaction (more localized compaction) of pigment particles. The device is also simple to fabricate because it can be made using low-cost roll-to-roll techniques. However, this switching is done with single-color particles switching between a clear and colored state and multiple color states can only be achieved by stacking layers which again limits the optical performance and resolution.

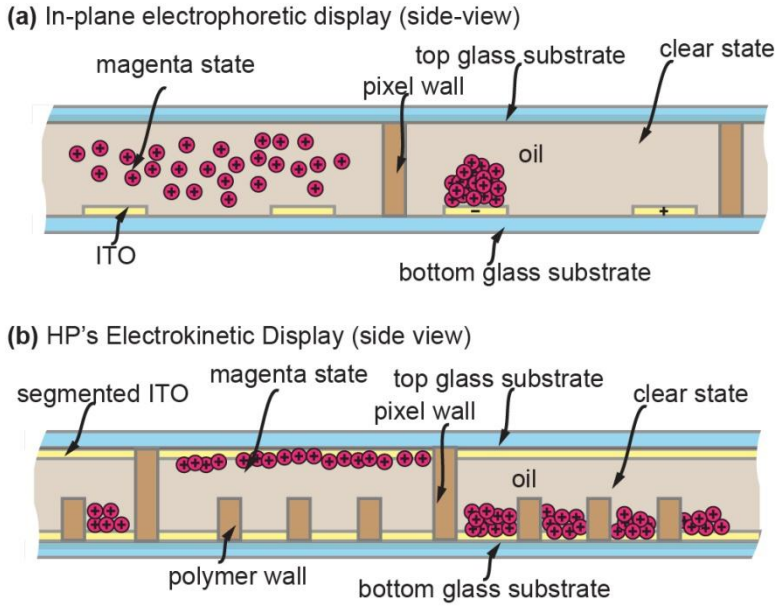


**Figure 1.8**: (a) In-plane electrophoretic movement (b) Hybrid electrokinetic movement

### 1.4 Research Aims and Outline

All the above-discussed approaches are techniques by which the electrophoretic pigments can be switched in a pixel, but lacking is successful switching the pixels in multi-spectral states using device structures that are commercially compelling. *This work will expand the applications and the capability of switching the electro-optical devices by implementing electrophoretic particle switching with multiple colored particles, so that greater spectral ability could be created in a single switch without significant increase in device or manufacturing complexity, and without large decreases in optical performance.*

Merck Chemicals Ltd. Has recently developed polymeric microparticles into which a dye, a charge (of either sign), and steric stabilizing surface modification can be built into the particle during synthesis [16]. They synthesized all the R, G, B, C, M, and Y colored electrophoretic dispersions and also created complementary colored dual particle pigment dispersions by pairing up R-C, G-M, and B-Y particles but with opposite charges on each colored particle type. These electrophoretic newly available particle dispersions are used to accomplish the central aims of this dissertation. Furthermore, this dissertation goes beyond basic demonstrations with these new particles, and demonstrates their switching in a compelling new device structure. The specific research aims of this dissertation are as follows:

***Aim 1: Characterize the fundamental behavior of two-particle electrophoretic inks in a variety of simple electrode configurations:*** for this aim, the two-particle inks can be modulated using in-plane electrodes (this dissertation) [16] using vertical electrodes, and using hybrid electrokinetic [14], [15] electrodes (this dissertation). The fundamental behavior of the two particle dispersions in each of these electrode configurations is to be evaluated first. Chapter 3 evaluates the functioning of the two particle dispersions and characterizes them for their various parameters that are important for designing more advanced device structures.

***Aim2: Create and explore a novel, advanced and grayscale capable e-Paper pixel using dual-color dual particle ink dispersions in an in-plane electrophoretic pixel structure to demonstrate the proof of concept of multi-spectral switching***. For this aim the behavior observed for a single particle electrophoretic display is used to research the nature of two color particle inks to create a unique electrophoretic behavior required for a biprimary color generation. By completion of this aim we will demonstrate the optical switching of pixels with four color-states including white (W) black (K), and two other colors, suitable for highly bright color e-paper performance. This aim will also accomplish high contrast ratio switching of colors in all the four states. Chapter 2 introduces the biprimary color system, a single layer color system which is theoretically capable of better reflectivity than the side-by-side RGBW system. The chapter 3 introduces the fundamental characteristics of these biprimary dual-colored dual-particle electrophoretic dispersions synthesized by Merck Chemicals Ltd. These particles are then implemented in in-plane device architecture to switch in 4 color states as explained. The device is characterized spectrally to evaluate the ink performance for a basic proof of concept to come up with advanced designs.

***Aim 3: Creating and exploring novel micro replicable hybrid electrokinetic device architecture for reflective displays and a smart window demonstrator, which uses dual particle ink dispersions for brighter displays and color-temperature tuning of smart windows.*** This aim successfully accomplishes a hybrid electrokinetic device structure that is highly micro-replicable, scalable and involves very few fabrication steps, that solves the problem of long-line electrode patterning which is a major drawback for the technology proposed by Philips research labs [17]. Chapter 4 introduces this structure and explores the spectral characteristics of the device for both the reflective and transmission analysis along with a COMSOL modeling of the structure to investigate the physics of the device operation.

The outline of the dissertation is as follows:

**Chapter 2**: Introduces the single layer color system called the biprimary color system that will be implemented with all the experiments in this dissertation. The advantages of the biprimary color system over traditional RGBW color system is explored and established both quantitatively and qualitatively.

**Chapter 3**: Implements the dual-particle biprimary colored dispersions in a number of electrode architectures to evaluate their functioning and behavior in such electrode architectures: in-plane, vertical and electrokinetic. The fundamental characteristics such as spectral properties, electrophoretic mobility etc., are evaluated to help project more advanced device performance. This chapter then implements the dual-particle biprimary colored electrophoretic dispersions in a simple in-plane electrode structure to prove the concept of multi-color switching. The simple structure will switch in four colored states: Black (K) White (W) and two complementary colors. This illustrates the basic proof of concept and paves the way for a more advanced architecture device presented in Chapter 4.

**Chapter 4**: This introduces a hybrid electrokinetic structure of device that implements the biprimary dual-particle dispersions in a structure that is highly manufacturable using even simple roll-to-roll processing. This structure is capable of switching the pixels in 4 states as per the in-plane structure demonstrates in chapter 3. The devices are demonstrated in modes needed for reflective displays (reflective) and smart windows (transmissive). For both the applications, this structure provides advanced performance or functionality that goes beyond that available with existing electro-optic switching techniques.

**Chapter 5**: This chapter will talk about the overall summary of the work presented and the future work that is possible based on this dissertation. Specifically, the chapter discusses potential work that could enable successful commercialization of the technology.

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# *Chapter 2: The Biprimary Color system*

## 2.1 Introduction to the chapter

This chapter introduces the biprimary color system which in one example implementation, can use dual color dual-particle dispersions that are complementary to each other (e.g. Red with Cyan, Green with Magenta, and Blue with Yellow). All these pairs consists of colors that are visible spectral complementary to each other. This chapter explores the motivation to create this color system and its advantages over the contemporary color systems like the side by side RGBW color filter arrays.

## 2.2 Background

We already discussed in chapter 1, the conventional switching of pixels available for various technologies switches between only clear and opaque states be it transmissive type or reflective type. A full color is generated only by placing a side by side RGB or RGBW color filter arrays. The other way that produces full color is subtractive, by stacking CMY layers one over the other. As already discussed in chapter 1, there are numerous disadvantages of employing these methods to generate full color switching or multi-color switching, and needed is a new method by which the number of filters or layers for generating color can be reduced without compromising color brightness. Ideally, this new method would also actually increase optical performance.

It has been previously stated that theoretically the best white brightness can be achieved by stacking the CMY subtractive color layers but this stacking comes with additional problems like low resolution with optical losses. The RGBW color filter array is the most prevalent way of generating the multi-color switching, but only provides color in 25% area of the pixel amounting to a maximum theoretical white state of 50%. Hence a new color system is introduced, known

as the biprimary color system that includes one color from RGB and one color from CMY so that they are visible light spectrally complementary of each other. For example, Red with Cyan as Cyan consists of Green and Blue; Green with Magenta (Red and Blue) and Blue with Yellow (Red and Green) demonstrates the pairing of the biprimary colors. The term ‘primary’ prefixed by ‘bi’ originates from unification of both the RGB and CMY primary color systems inside a single pixel. A typical biprimary system is now illustrated in Figure 2.1, and will first be compared to the conventional RGB and RGBW color systems..

The most commonly used color system is side-by-side RGBW color pixels, and provides a maximum white reflectance of 50% as calculated from the average of three RGB sub-pixels that reflect 1/3rd of the visible spectrum and a fourth white sub-pixel (Figure 1.1). The fourth white sub-pixel is only added to boost the overall reflectance. The RGBW system has a color-fraction (CF) of 25% because at maximum, a saturated color such as red can only be displayed at 1/4th of the total pixel area. The term CF is used in place of color gamut, color saturation, or other terms because it is easy to calculate and compare across various color systems. Actual color gamut, and human perception of color, is non-linear with CF. Hence, RGBW can at best provide maximum theoretical reflectance of ~50% and CF of~25%, and in practice, the performance is typically much lower due to optical pixel inefficiencies [1].

The bi-primary color system [1][2] can be of two forms; a non-mixing, and a mixing system shown in Fig. 1. Both bi-primary color-systems will display two complimentary primary colors inside each sub-pixel as explained. Each RGB primary color is paired with its CMY compliment in three sub-pixels. The non-mixing bi-primary (Fig. 1a) will have the two colors displayed side-by side in the pixel, but not mixed, and when no-colorant is displayed in the pixel the color is black. For example, red and cyan could each occupy half the sub-pixel area but cannot overlap. Since they cannot overlap, they cannot create black. Hence a 3rd black colorant or black background is required if we need to create a black pixel. Pixel technologies that could utilize

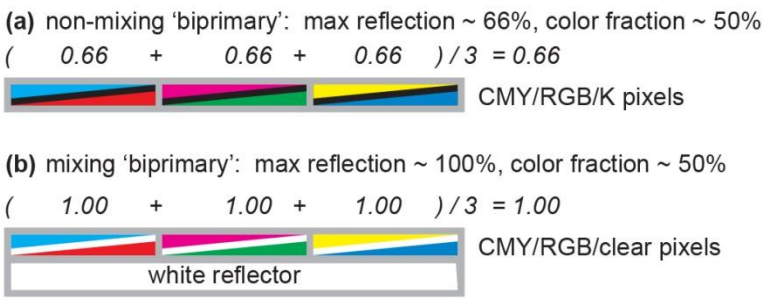


**Figure 2.1**: (a) Side-view of non-mixing biprimary pixels (b) side-view of mixing biprimary pixels (reproduced from reference [1])

non-mixing bi-primary include Electrowetting [4] and Electrofluidic [5] where colorants are moved in a fluid (not through a fluid). On the other hand, the *mixing* bi-primary (Fig. 1b) allows the complimentary colors to mix (overlap) and therefore create black, or the colors can be cleared up from the pixel area they can reveal a white background. Pixel switching technologies that could utilize mixing biprimary include electrophoretic (vertical [3] and horizontal in-plane [6][8], or combinations thereof such as Elecrokinetic [7]). Other technologies such as liquid crystal or MEMs may also be capable of using non-mixing or mixing bi-primary function, but currently lack the ability to do so. It is evident that bi-primary is best suited for colorant-transposition technologies. This is an advantage, because colorant transposition would be the best option for pixel switching in multi color states and exhibit some of the highest reflectance and CF as already established in Chapter 1.

The maximum reflection and CF performance for bi-primary is best understood by Fig. 2.2, which contains top view pixel illustrations and example calculations for display of W, R, and C colors. First, consider W (Fig. 2.2a). The non-mixing bi-primary would display CMY for a theoretical reflection of 67%, whereas the mixing bi-primary could clear the colors from the pixel for a theoretical reflection of 100%. The mixing bi-primary therefore provides 2X the white reflectance of RGBW color-filtering. Next example, consider reflectance for displaying red (Fig.

2.1); both the mixing and non-mixing bi-primary methods would display RMY for a reflectance of 55% (33% from R, and 67% from each M and Y). The CF is also improved compared to RGBW. R contributes 100% CF and M&Y 25% each. The CF for M&Y is 25% because each is comprised of half R (50% CF) but also a non-red color (B, G) which reduces their CF by a factor of two. The resultant CF is 50% which is ~2X the CF for RGBW. The case of displaying C also illustrates the advantage of biprimary. The non-mixing biprimary can provide a reflectance of 44% and a very high CF of 67%. A better CF leads to a better saturation of colors, which in turn leads perceptively to higher brightness. The overall reflectance can be further be boosted for CMY and RGB colors when the mixing bi-primary pixel can sacrifice some CF. In Fig. 2.2c, the mixing bi-primary has two pixels which are incompletely colored (showing color over half their area with W in the other area), thereby boosting reflectance to 67% while maintaining a CF of 50%. This CF, even though reduced, is still double the value achieved for RGBW. All other color combinations, including K, can be achieved using the bi-primary color system.

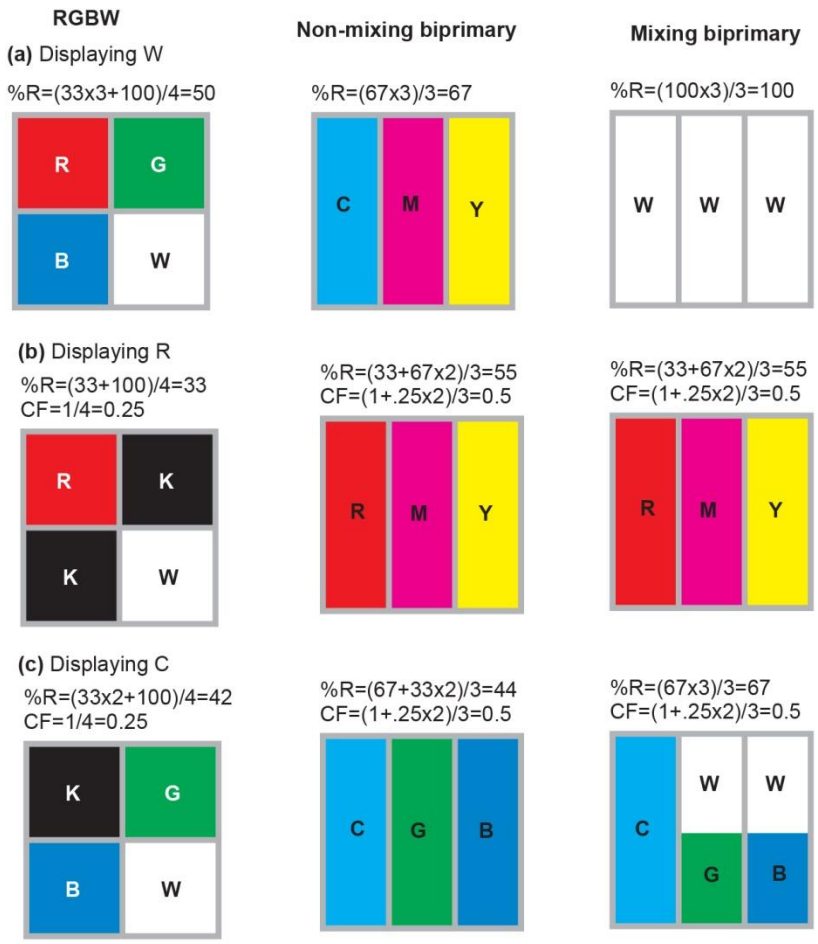


**Figure 2.2**: RGBW, non-mixing biprimary and mixing biprimary showing colors for (a) W (b) R and (c) C (reproduced from reference [1])

Diagrams and calculations for additional colors are not reviewed because their calculations are similar to those for W, R, and C.

### 2.3 Application of Biprimary color system in electrophoresis

As already been discussed, the suitable method of achieving multispectral switching inside a single pixel can be by using colorant transposition for which electrophoretic displacement of particles sounds most feasible. Also, the biprimary color system can successfully utilize this

method through the mixing biprimary type, where two different colored particles can be mixed in dispersion and then moved such that different color states can be achieved. The different types of electrophoretic pixel switching as already discussed in the previous chapter as vertical or in-plane horizontal type. For switching with a vertical type as already discussed in chapter 1, the vertical dimension transposes the color particles that need to be displayed, and for utilizing a mixing biprimary type, the two different colors can be displayed just as shown for a B/W in Figure 1.5 for a typical E-Ink display. A K state can be displayed by mixing them but a third white particle would be required to create the W state. This system is not ideal due to its need for a three particle system.

The most suitable way of utilizing the advantage of the mixing biprimary with electrophoretic movement of particles is by moving them in plane such that when the two colors are mixed they represent black, they can be cleared up to obtain a clear pixel, and each color can be displayed while the other one is still compacted to obtain the two different color states. A similar approach is described in Figure 1.6. This approach will be explored in detail in the subsequent chapters.

**2.4 Comparison of Biprimary color system versus RGBW**

The Figure 2.2 explains the theoretical color fraction (CF) and reflectance of W, R and C and also demonstrates the sub-pixel colors for each color compared with the RGBW. As shown in the calculation, the biprimary colors theoretically boost the reflectance and the CF by approximately a factor of 2. An artificial pixel layout is shown in Figure 2.3 [9] that demonstrates the biprimary colors and RGBW colors in details.

For each color shown in the figure, a zoom in inset diagram is shown for the three sub pixels comprising a single biprimary color pixel. It is important to note that the artificial pixel layouts a black space amounting to 20% of the area in order to mimic a reasonable fill factor for a real pixel. The pixels in the Figure are provided in as drawn form and also in blurred form (Adobe

Photoshop, Gaussian Blur 9.0) to mimic visual appearance at a normal viewing distance. Figure b shows the same for RGBW pixels

A more quantitative colorimetric analysis is performed in the data plot shown in Figure 2.4. The digital image files of the artificial pixel layouts of both the biprimary and RGBW is used to extract the theoretical La*b* points using a digital color meter. The gamut area has been calculated for SWOP (specifications for web offset printing), biprimary and RGBW for both the plots using the following equation [10].

$$A = \frac{1}{2}\left|(a_1^*\text{-}a_2^*)\,(b_1^*+b_2^*)+\,(a_2^*\text{-}a_3^*)\,(b_2^*+b_3^*)\ldots\ldots\ldots\,(a_6^*\text{-}a_1^*)\,(b_6^*+b_1^*)\right|$$

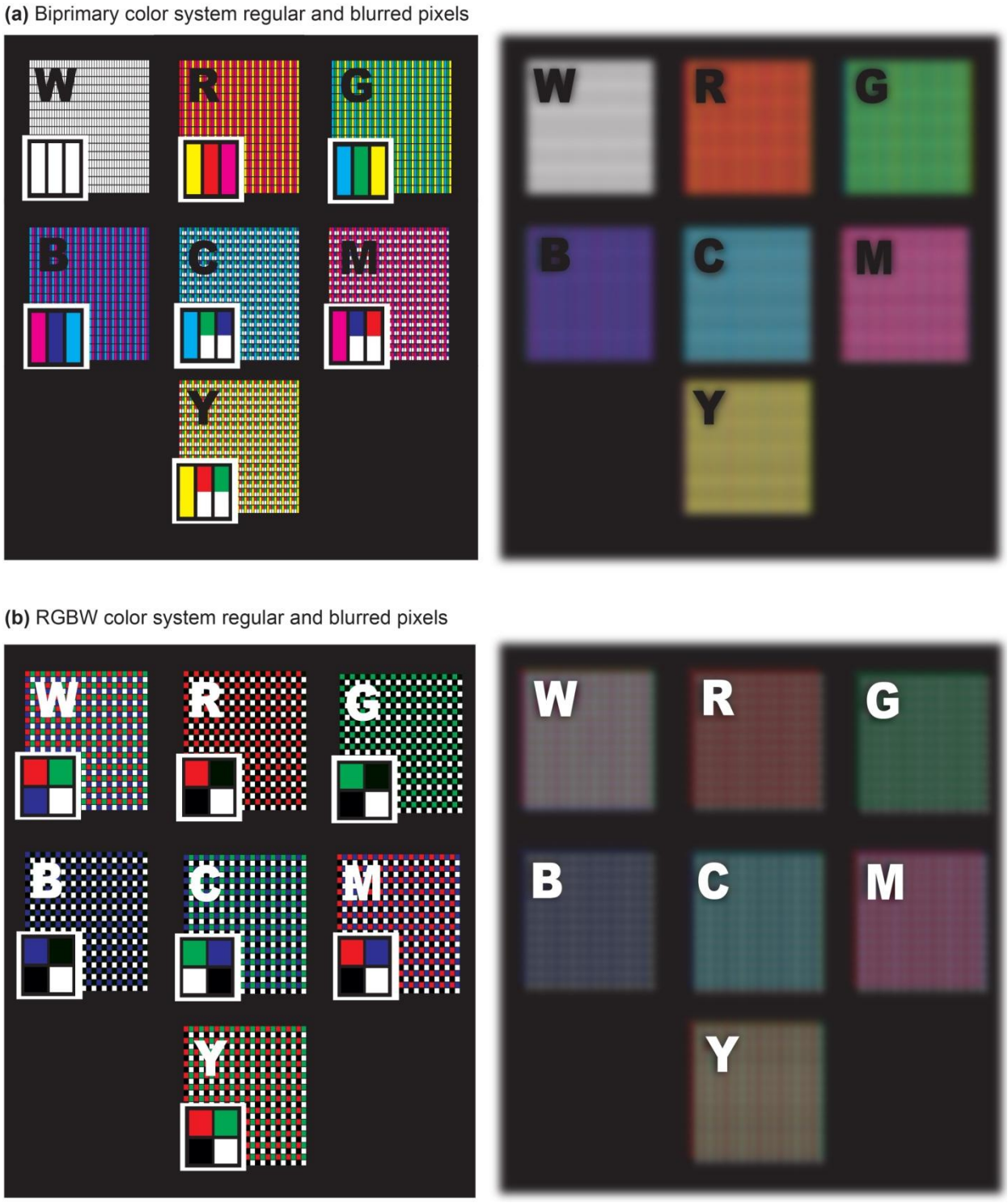


**Figure 2.3**: RGBCMYW colors in (a) Biprimary color system showing regular and blurred (b) RGBW color system showing regular and blurred (reproduced with permission from ref [9])

As can be seen from the colorimetric simulation and also calculated from the plot the theoretical performance is found to be ~33% of the SWOP area for the biprimary while the RGBW is found to be ~4.5% of the SWOP area.

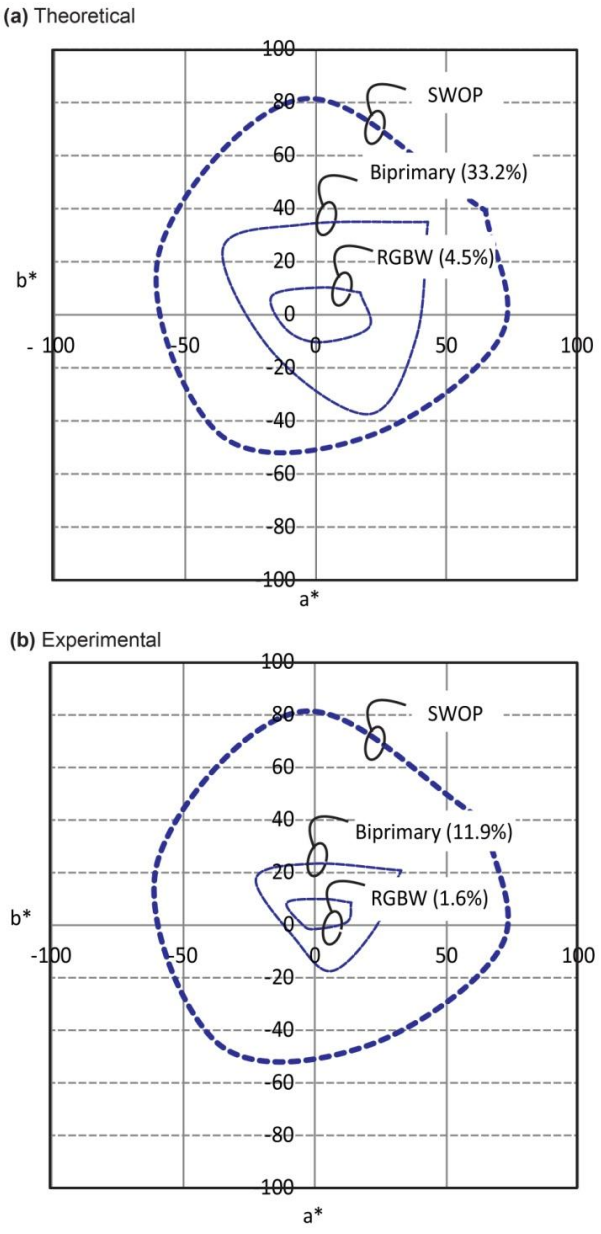


**Figure 2.4**: (a) Theoretical color space mapping (b) Experimental color space mapping, both compared to SWOP color space represented by the outer circle (reproduced with permission from ref [9])

The blurred pixels are also printed on a regular printing paper (80% reflectivity) using a regular HP Color LaserJet CP4525 color printer. This provided an example of color optimized pixels, which at some point could also be duplicated in the biprimary pixels. This example only used in a conventional reflector of 80% and a more sophisticated reflector can boost the reflection further. The printed pixels are then measured by a Minolta CS colorimeter and a $D_{65}$ Illuminant. Although experimentally measured color-space is reduced from the theoretical one, they still comprise of a larger fraction in the SWOP color space than their RGBW counterparts. Hence this presents the superiority of the Biprimary colors over the RGBW color system both quantitatively and qualitatively.

**2.5 Conclusion**

We introduced in this chapter a color system called the Biprimary color system which has the potential to be the next generation single layer color system that can be implemented with large area display applications, switching pixels in multi-spectral states. The best approach was found to be using electrophoresis as already explained in chapter 1, using the biprimary dual-particle dispersions. Potential improvements in color performance reflection and brightness can be obtained theoretically up to 2X over the RGBW system. The simulation and data plots supports this argument that biprimary is potentially a higher performance color system than the side-by-side RGBW. In the next Chapter, we will discuss how to implement these biprimary dual-particle dispersions to switch pixels in multi-spectral states and explore their benefits and drawbacks in subsequent chapters.

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# *Chapter 3: A first demonstration and analysis of the biprimary color system for reflective displays*

## 3.1 Introduction to the chapter

In this chapter a new biprimary color system is demonstrated for single-layer reflective displays, capturing much of the improved color performance of multilayer displays while potentially maintaining single-layer display advantages in high resolution and faster switching. Electrophoretic pixels were operated with dual particle complementary-colored dispersions such as green/magenta (G/M). Using simple interdigitated three-electrode architecture, four colored states (KWGM) were achieved with a preliminary contrast ratio of 10: 1. Furthermore, biprimary ink dispersions were shown to be functional in a more advanced electrokinetic pixel structure. A full-color biprimary pixel contains three complementary sub pixels (G/M, B/Y, R/C), and the requisite electrophoretic ink dispersions were also formulated and spectrally characterized in this work. The theoretical color space mapping in chapter 2 has already confirmed that the biprimary concept provides twice the brightness and twice the color fraction compared with the conventional RGBW sub pixel approach, and that the biprimary concept can approach performance close to that of magazine print (Specifications for Web-Offset Print). This Chapter deals with the actual demonstration of that concept and the projected future prospects of this method.

## 3.2 Background

Reflective displays, often referred to as ‘electronic paper’ or e-paper, have for at least a decade, been assumed to be the future technology for sunlight-readable, low-power, reduced weight, and the preferred route to achieve flexible or rollable displays.[1] In support of this assumption, video-rate e-paper technology is now achievable, including Electrowetting [2],[3] and Micro

Electro Mechanical Systems (MEMS) technologies.[4],[5] However, of the dozen or more technologies that exist, none are able to provide bright color operation without moving toward multi-layer Cyan-Magenta-Yellow (CMY) color generation, and therefore having to accept significant compromises in switching speed [6],[7] or pixel resolution.[8–10] Therefore, faster switching speeds or higher pixel resolutions are typically relegated to lower-performance color systems such as side-by-side RGBW pixels, which can only display saturated color at 25% of the display area (color fraction (CF) = 25%).[11] What is needed, and has not been yet demonstrated, is a color system, which merges the cost, resolution, and switching speed advantages of single-layer color-additive displays, with the improved color performance of multi-layer color subtractive displays. A new paper written by our research group tries to solve this existing problem [11]. Demonstrated here is a new biprimary color system [12], as we introduced in the previous chapter, which provides a doubling of both color and brightness and is able to do so using a highly desirable single-layer implementation. As already explained, the term ‘primary’ prefixed by ‘bi’ originates from unification of both the RGB and CMY primary color systems inside a single pixel. As shown in Chapter 2, each sub pixel can be dual-colored with one of the RGB primaries and its complementary color in the CMY primaries. W is achieved by clearing the colors, K by fully mixing them, and bright colors such as R achieved by activating the sub pixel colors that most strongly contribute to R, for example, a display of RMY where M and Y themselves are half-red in their spectra. The experimental demonstration in this work utilizes electrophoretic pixels and two-particle two-color ink dispersions [13]. Pixel fabrication and characterization is performed for the G/M sub pixels and the potential of inks for the other sub-pixels (R/C, B/Y) are analyzed using reflection analysis. With the G/M inks and simple interdigitated three-electrode architecture, all four states of KWGM can be achieved with contrast ratios of up to 10:1. A more sophisticated electrokinetic pixel structure (faster, two-electrodes) is also demonstrated for the G/M ink. A theoretical color-space analysis and display simulation is provided in the previous chapter, which visually shows the qualitative doubling of

brightness and CF as compared with the conventional RGBW approach. The predicted biprimary performance is close to that of color-quality standards for magazine print (Specifications for Web-Offset Print or SWOP) that has also been documented in the previous chapter. Although these are preliminary results, they confirm that biprimary pixels can be fabricated and operated under the basic fundamentals for biprimary color.

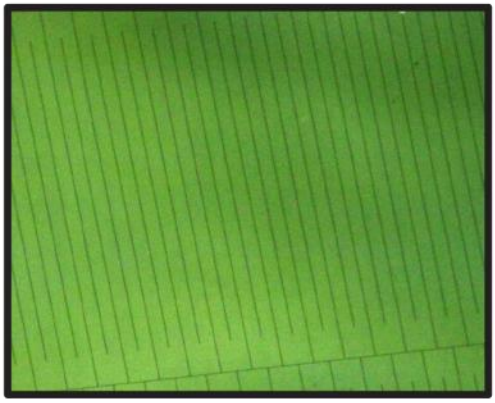
### 3.3 Biprimary experimental demonstrations

#### 3.3.1 Biprimary dual particle dispersions

The dual particle ink dispersions, based on dyed polymer microparticles [13] used in the device are a key enabling material for the biprimary color system. It has been demonstrated by Merck (known as EMD in North America) that the particle design, color, size, charge, and surface functionality can be independently tailored with the use of suitable dyes to realize any combination of two colored particles including those from the subset of RGBCMY. Particle synthesis enables covalent combination of dye, charging components (of either sign) and a steric stabilizing surface modification.

Merck Chemicals Ltd. Has recently developed polymeric microparticles into which a dye, a charge (of either sign), and steric stabilizing surface modification can be built into the particle during synthesis [13]. The steric stabilization provides long hydrophobic tail (like alkanes) that extend into the hydrophobic oil that the particle is dispersed in. The steric stabilization is special, though, because the hydrophobic tails also prefer not to interact with each other. This also prevents particles from agglomerating or settling with each other. The polymer phase provides the correct charge and the dye phase provides the correct color. They synthesized all the R, G, B, C, M, and Y colored pigments and the complementary colored dual particle pigment dispersions by pairing up R-C, G-M, and B-Y particles.

**(a)** A device showing OFF or combined B/Y (K) state



**(b)** A device showing ON or compacted B/Y (W) state

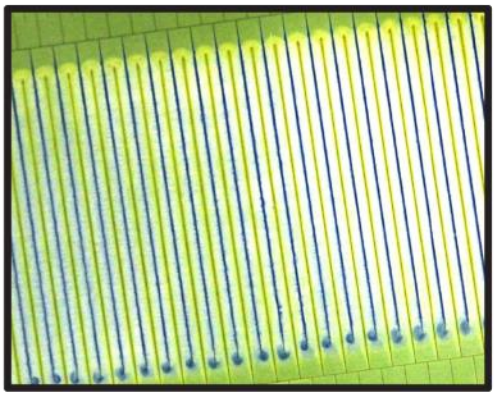


**Figure 3.1** (a), (b) a device operating B/Y dispersion in a simple interdigitated electrode layout

The preliminary tests were conducted on B-Y dual particle dispersions on a simple parallel interdigitated electrode test cell as illustrated in Figure 3.1. The electrodes are placed at a distance of 300um from each other and a 70V DC voltage source is applied. In this test Y and B were negative and positively charged respectively. A simple clear (W) state (ON) and mixed state (K) was achieved (OFF). This demonstrates that the dual color/dual particle dispersions can be reversibly actuated without agglomeration. The reflection data shows the achievable B/Y reflection and their combined theoretical K state.

The table below shows the critical parameters for all the color dispersions synthesized by Merck. Thus based on the preliminary results, it can be claimed that dual color/ dual particle

| Sub-Pixel Pair | Particle-Size (nm) | Polydispersity | +Zeta (mV) | -Zeta (mV) |
|---|---|---|---|---|
| Red | 381 | 0.09 | 49.3 | 63.9 |
| Cyan | 549 | 0.09 | 68.3 | 65.2 |
| Green | 826 | 0.44 | 76.5 | 87.6 |
| Magenta | 690 | 0.15 | 59.5 | 73.2 |
| Blue | 311 | 0.07 | 89.5 | 24.8 |
| Yellow | 779 | 0.17 | 51.6 | 70.9 |

**Table 3.1**: The properties of all the RGBCMY particles as synthesized by Merck

dispersions can be implemented in a more complex pixel-like architecture to control each type of color separately, and in turn be able to switch pixels into multiple spectral states.

In this chapter, work is focused on the green/magenta dispersions. In this specific work, green particles are negatively charged, whereas the magenta particles are positively charged. Electrophoretic mobilities for the particles were measured and reported in a later section of this paper.

### 3.3.2 Device Fabrication

In this chapter, a 3 electrode in-plane electrophoretic pixels were demonstrated, with the 3 electrode system utilized in most of the experiments. The 3 electrode system (Fig. 3.2) is simpler to fabricate and increases the maximum optically active area, but unlike the 4 electrode system, it requires a clearing or 'reset' state in between color changes. There are two moving electrodes (ME1 and ME2) and one gating electrode (GE), all fabricated 'in-plane' (on the same substrate).With 4 electrodes, another gate GE2 could be added adjacent to ME2 (not shown). The electrodes are made from transparent In2O3:SnO2 (ITO), patterned by wet etching and

photolithography. The test device is assembled with a transparent top-plate, and the biprimary ink is dosed similar to the 1-drop filling technique used in liquid crystal display manufacturing. Once the device is assembled, the electrodes ME1, ME2, and GE are operated individually with three-way switches to enable 0, +32.5,-32.5 V DC voltage control. When tested in reflective mode, a rear reflector is required. For higher resolution pixels and to minimize light-out coupling losses [14], the rear reflector should be as close to the pixels as possible but also separated from the pixels by a low-refractive index layer or air-gap.

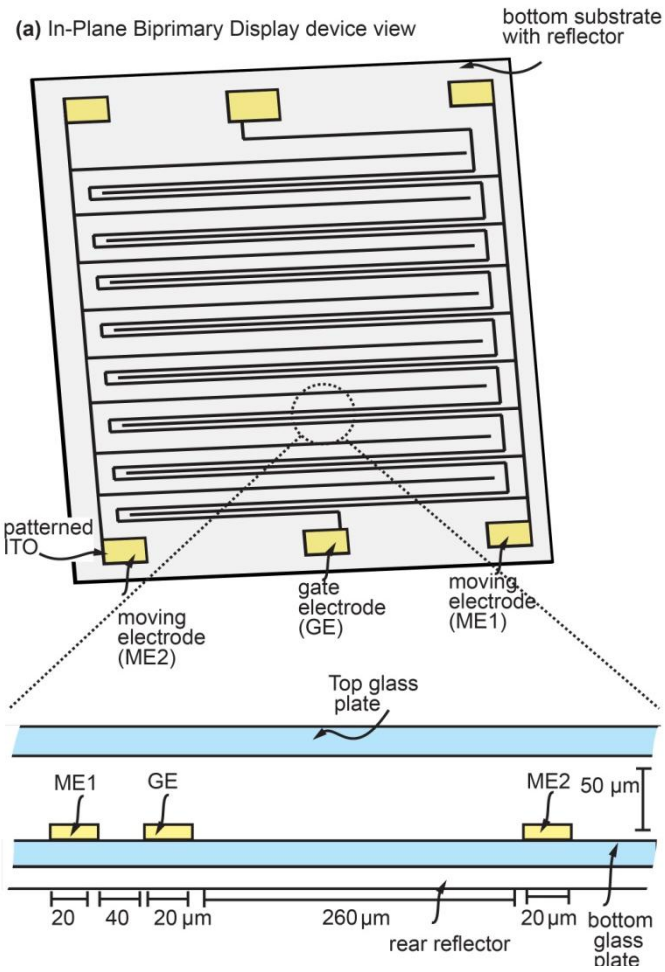



**Figure 3.2**: A basic 3 electrode in-plane top view and cross-sectional view respectively

In this work, the rear reflectors were fabricated by coating a 25-μm thick polyethylene sheet with light-scattering (diffusing) barium sulfate powder (BaSO4), mixing with a small amount of organic binder, and placing that sheet on the top of a 99.8% reflective 3-M Vikuiti$^{TM}$ Enhanced Specular reflector (ESR)film.

### 3.3.3 Device Operation

In-plane electrophoretic displays work on the principle of using an electric field to move charged pigment particles towards or away from the viewable area in each pixel (colorant transposition). The operation of the biprimary color dispersions are illustrated in Fig. 3.3 and photographs of K, W, G, and M states are provided in Fig. 3.4.

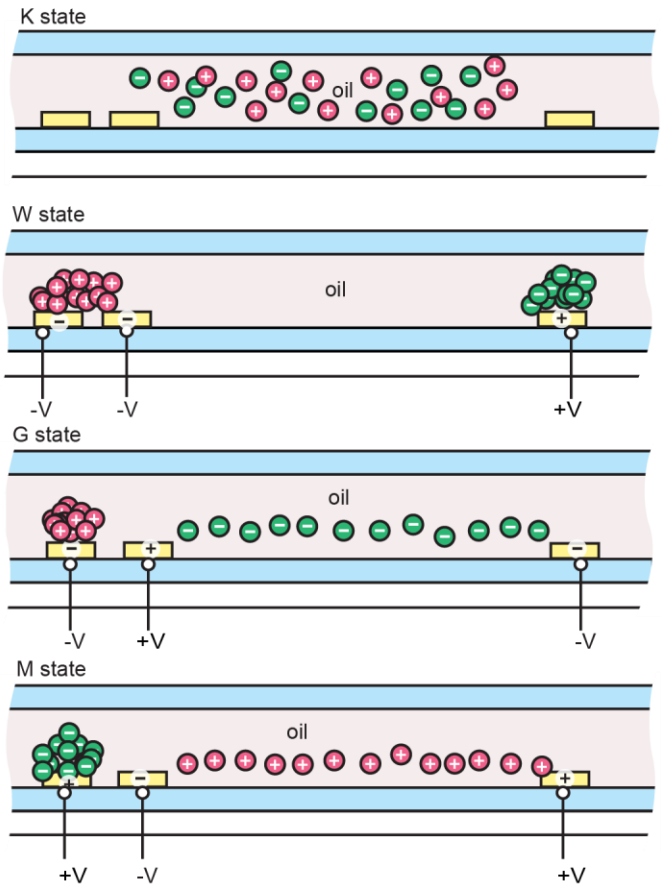



**Figure 3.3**: Voltage switching for all the 4 K, W, G and M states

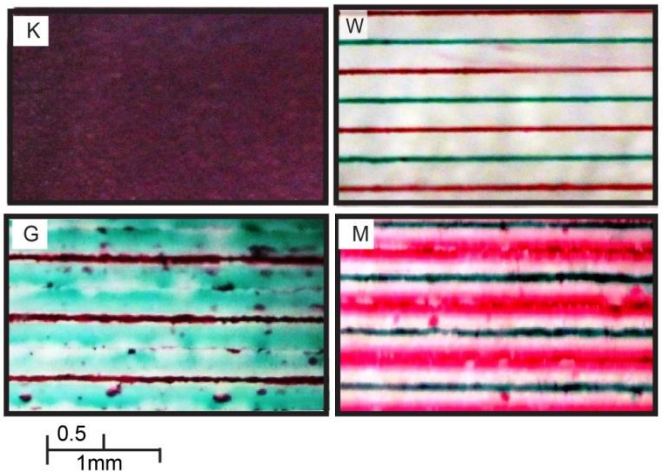



**Figure 3.4**: The photographs of all the K, W, G and M states

Using pulse-width modulation of particle spreading or other techniques, grayscale can be achieved [15] but was not demonstrated in this work. In this work, each of the colored states was achieved using fully colored or cleared states. The voltage sequences were as follows:

***Black state*** *(K)*: Firstly, the pigments are all compacted onto electrodes by setting ME1 = +32.5 V and ME2 =-32.5 V (with a net potential difference of 65 V) and GE = +32.5 V. Next, all the electrode polarities are reversed for a duration of ~7s to spread the pigment particles (incomplete movement across the pixel) then voltages are removed allowing the particles to remain in a fully mixed (black) state.

***White state*** *(W):* Next, the voltage is applied as shown in Fig. 3.2b with ME1 = 32.5 V, ME2 = +32.5 V, GE = 32.5 V, and the pigments are compacted onto the electrodes, revealing the white reflector at the background.

***Green state*** *(G):* After obtaining W, voltages are set as ME1 = 32.5 V, ME2 = 32.5 V, and the GE electrode is switched to +32.5 V, which (1) confines the M pigment compacted on ME1; and (2) spreads the G pigment across the viewable area. After ~10 s, the G spread state is

achieved, and the voltage between ME1 and GE is then set to a value of 10 V to sustain M compaction on ME1, and 0 V between GE and ME2 to sustain the spread of G pigment.

***Magenta state*** *(M)*: M is obtained by again first setting the W state, and using the opposite polarities as were described earlier for setting the G state. The diffused spectral reflectance data of these states were measured and are plotted in Fig. 3.5. Biprimary switching behavior is seen in the plots, but is also non-ideal in spectral performance as the G pigment does not fully suppress M reflection and M pigment likewise does not fully suppress G reflection. These particle dispersions utilize typical dyed polymer microparticles from Merck and are not optimized for biprimary operation. Therefore, improvements in maximum reflection, color reflection, and in black states are all expected in future work

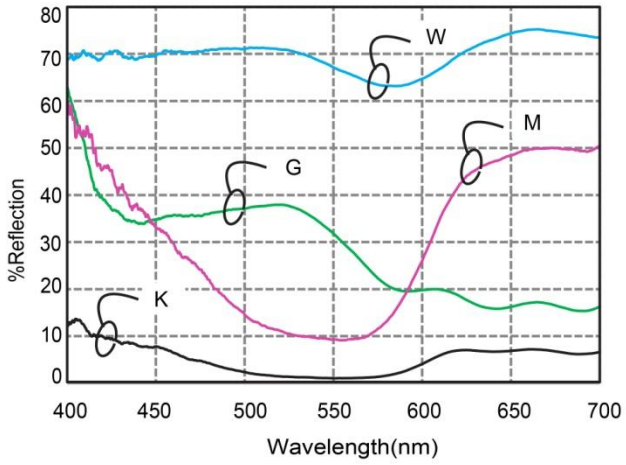



**Figure 3.5**: The reflection data of all the 4 KWGM states measured in the device

### 3.3.4 Electrophoretic mobility and speed

Electrophoretic mobility for the dual-particle green-magenta dispersion ink was tested in a simple two interdigitated electrode test cell (ME1 and ME2 only, no GE, Fig. 3.2). These electrodes are 20 μm wide and were spaced at 300 μm distance from each other, and the applied voltage was 70 V. The apparent electrophoretic mobility of the particles was then calculated using Image J analysis of video of the moving particles. The speed of the particles was measured every 50μm distance, and the electrophoretic mobility constant (μ) is calculated using the common formula:

$$\mu = \frac{E}{V} \text{ cm}^2/\text{V-s}$$

Where v is the velocity of the particles and E is the applied electric field. The plot in Fig. 3.6 shows the trend of electrophoretic mobility of both the green and magenta particles. Two important observations can be made. Firstly, the average mobilities are in the range of 3 to 6 × $10^{-6}$ $cm^2$ /V-s range, which is an order of magnitude lower than the best electrophoretic dispersions in existing commercial products. Achieving mid $10^{-5}$ $cm^2$ /V-s mobilities, which is 10 times the calculated mobility value, and small electrode spacing (~tens of μm) is essential if near-video speed switching is to be achieved (tens of ms). Secondly, as can be seen in Fig. 3.6, the velocity of the particles is not constant, and therefore the apparent mobility changes. The apparent mobility decreases as particles get closer to their final destination electrode, implying that the particles provide some repulsive force as they accumulate and start to internally screen the applied electric field. This effect is important, because when scaling the pixels to higher resolutions, the switching speeds will be slower than that predicted by the maximum mobility. Based on the data in Fig. 3.6, with each 50 μm distance decrease in electrode pitch, there is roughly a reduction of ~20% in the electrophoretic mobility. The electrophoretic mobility of the other pairs is also found to be of similar order hence not included in this chapter separately.

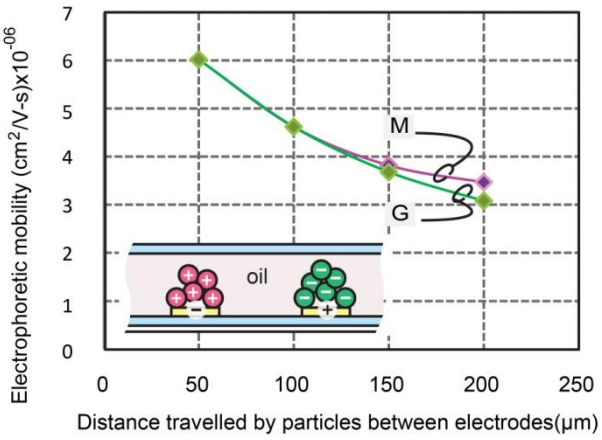



**Figure 3.6**: Plot of Electrophoretic mobility versus distance traveled by color particles between electrodes

Theoretically the electrophoretic mobility values will depend upon the values of zeta potential, dielectric constant of the medium and the viscosity of the dispersion medium. The following Huckel approximation is used for thicker double layer around the particles with size <1um:

$$\mu = \frac{2}{3} \times \frac{\zeta \varepsilon}{\eta} \times F(Ka)$$

Where ε is dielectric constant of the medium given by $\varepsilon_0\varepsilon_r$, where $\varepsilon_0$ is the permittivity of free space, $\eta$ is the viscosity and $\zeta$ is the zeta potential and *F (Ka)* being a function of particle radius *a* and inverse of Debye length $K^{-1}$.

The dispersion medium is dodecane oil with a viscosity of 1.34 mPa s, the relative permittivity is 2.0, if the zeta potential for green particles and magenta particles are 76.5mV and 73.2mV respectively (values from table 3.1), then the theoretical mobility values for green particles are $6.637 \times 10^{-6} cm^2/V\text{-}s$, and for magenta particles it is $6.35 \times 10^{-6} cm^2/V\text{-}s$, when assuming *F (Ka)* is 1 for simplicity [13]. Similarly the theoretical values of the other particles can be calculated separately.

### 3.4 Electrokinetic pixel demonstration

The green-magenta dual particle dispersion was also tested in a basic electrokinetic device (EKD) structure provided by Hewlett Packard (HP) Corp. A far more advanced version of this device will be presented in the next Chapter. The device cross-sectional structure is illustrated in Fig. 3.7. The bottom plate of the device assembly consists of a sheet Indium Tin Oxide (ITO) electrode, onto which hexagonal pixel structures are formed. The regular hexagonal pixels have an array of pits. The top plate is a transparent glass plate with a whole area ITO coating, kept at a channel height equal to the side walls of the hexagonal pixels. Exact dimensions are proprietary to HP. Figure 3.7(a) shows a Scanning Electron Microscopy (SEM) image of this EKD pixel, which is fabricated by a roll-to-roll manufacturing platform. The principle of operation for electrokinetic pixels includes both an out-of-plane (vertical) and an in-plane (horizontal) movement of the pigment particles. The electrokinetic effect can hence be called a hybrid between in-plane and vertical electrophoretic effects.

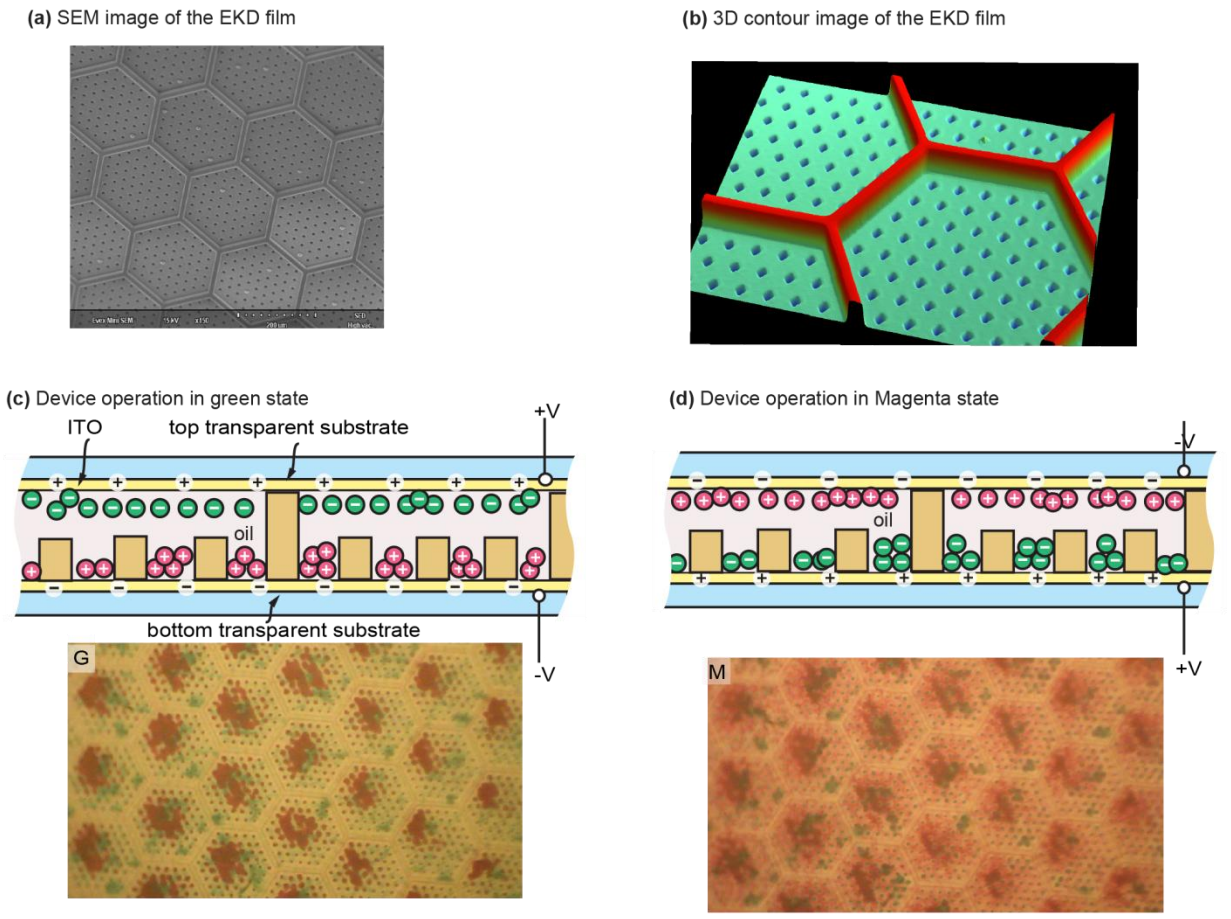



**Figure 3.7**: (a) SEM of Hp's electrokinetic film (b) 3D contour image of the EKD film (c) device operation in G state and (d) in M state

The electrokinetic device was driven with 20 V. Less voltage is needed compared with the in-plane electrode devices because the distance between the electrodes is >10× smaller. Two colored states were demonstrated as follows as in Fig 3.7 (c) and (d)

***Green state*** (G): To obtain G, the bottom plate is switched to -10 V, and the top plate set to +10 V, which pulls the M pigment down and hides it in the micropits, and which pulls up and spreads the G pigment.

***Magenta state*** (M): To display M, the bottom plate is switched to +10 V, and top plate switched to –10 V, which compacts the G pigment and spreads the M pigment.

The switching time of the dual-particle dual-color dispersions in the EKD pixels was found to be ~700 ms, which compares well to the HP's single-color ink, which switches <500 ms . The color performance is lacking, as the dispersions are not yet optimized for EKD operation. These results do show that biprimary EKD operation is possible. However, for color grayscale to be achieved, a gating electrode will need to be added to the pixel structure. There could be some applications that do not require a gating electrode. For example, consider a blue-yellow dispersion utilized for simple signage, and capable of displaying blue, yellow, or black, using only a single pixel structure and only two electrode contacts per pixel.

### 3.5 Predicted spectra for full color operation

In this work, G/M pixels were fully characterized, and other dual-particle dual-color dispersions also are available to satisfy the remaining C/R and B/Y sub-pixels in a biprimary display. These particles have similar mobilities, so the performance parameter of greatest interest is their spectral performance: if the spectral transmittance of the pigments is known, then the reflectance of the display is the product of the white background reflectance and the pigment transmittance squared. Figure 3.8 lists the reflection spectrum data (specular excluded) for each biprimary combination. In each plot, the two colors are measured for their reflection individually in a 50μm channel between two glass slides, and the K state is obtained by calculating the reflection of the combined state using the following equation where X and X' are the two complementary colors, the equation is:

$$\%R_K = \frac{\%R_X, X\ \%R_X}{100}$$

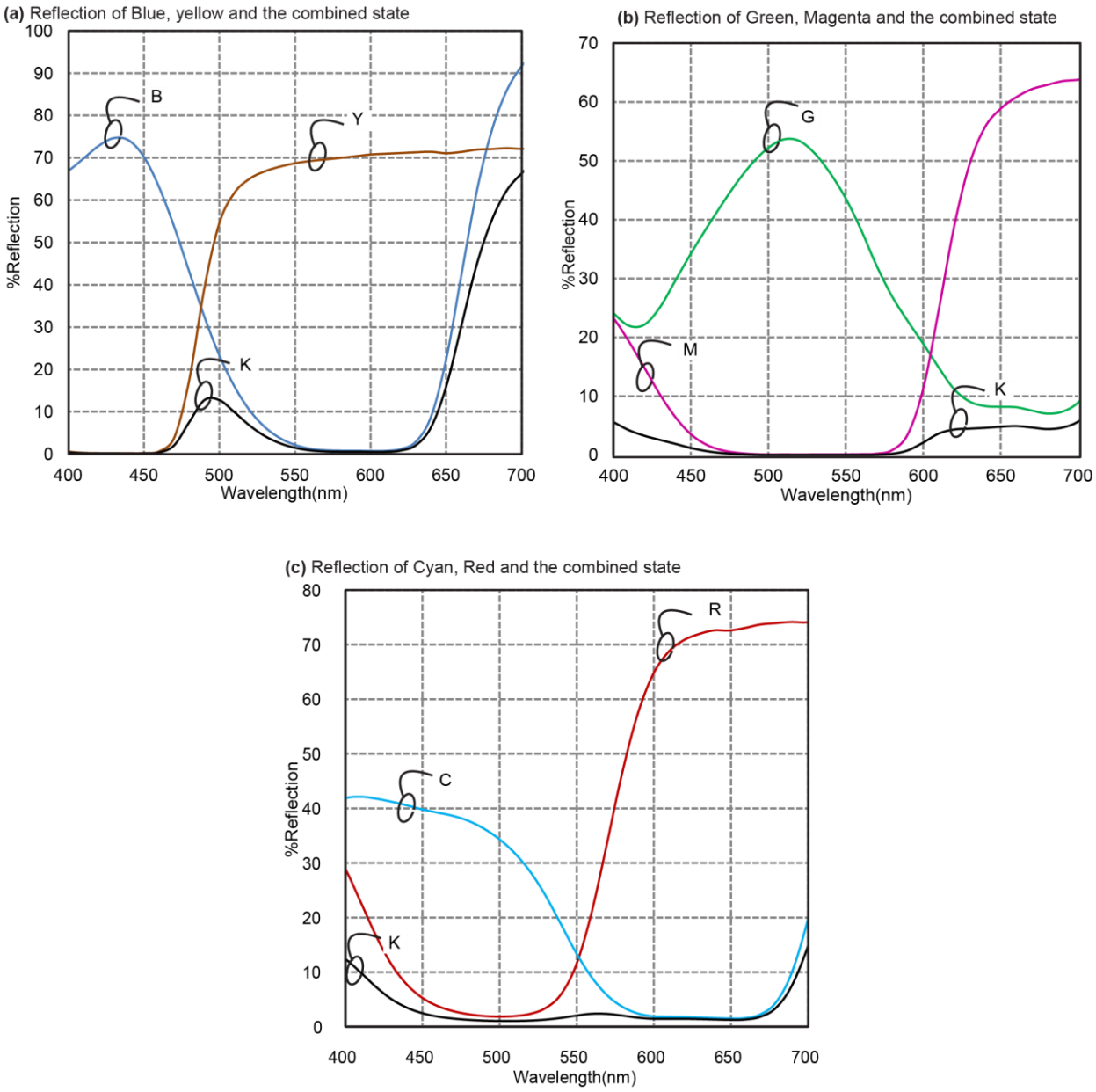



**Figure 3.8**: Reflection plot of all the 3 biprimary pairs and the combined theoretical K state for (a) B/Y (b) G/M and (c) R/C

Again, the particles are not optimized for biprimary operation and the maximum reflection values are below what is theoretically possible due to the spectral absorbance of the pigments, spatial distribution of the pigments and the total internal reflection at the display surface. Of particular interest in the spectral data is the black state. Strong black inks that are not based on carbon-

black typically require five or more colorants (dyes, pigments) to achieve uniform light absorption across the visible spectrum. Therefore, as expected and as can be seen in Fig. 3.7, there are small portions of the reflection spectrum, which limit the black state for the preliminary two-colored particle dispersions of this work. Figure 3.9 provides a comparative analysis of the theoretical K states of all the three biprimary pairs (C/R, M/G, B/Y), and their 'luminous reflectivity' obtained by multiplying the %R with the phototopic lumen/watt equivalent for each wavelength [17]. This is a better measure than just raw-reflectivity and reveals that the blue-yellow dispersion would exhibit the poorest black-state as perceived in terms of brightness by the human-eye.

The color space comparison between biprimary and RGBW color systems already established in the previous chapter provides qualitative and quantitative comparison between the two color systems with biprimary standing out in both aspects being a much better option as a single layer color system.

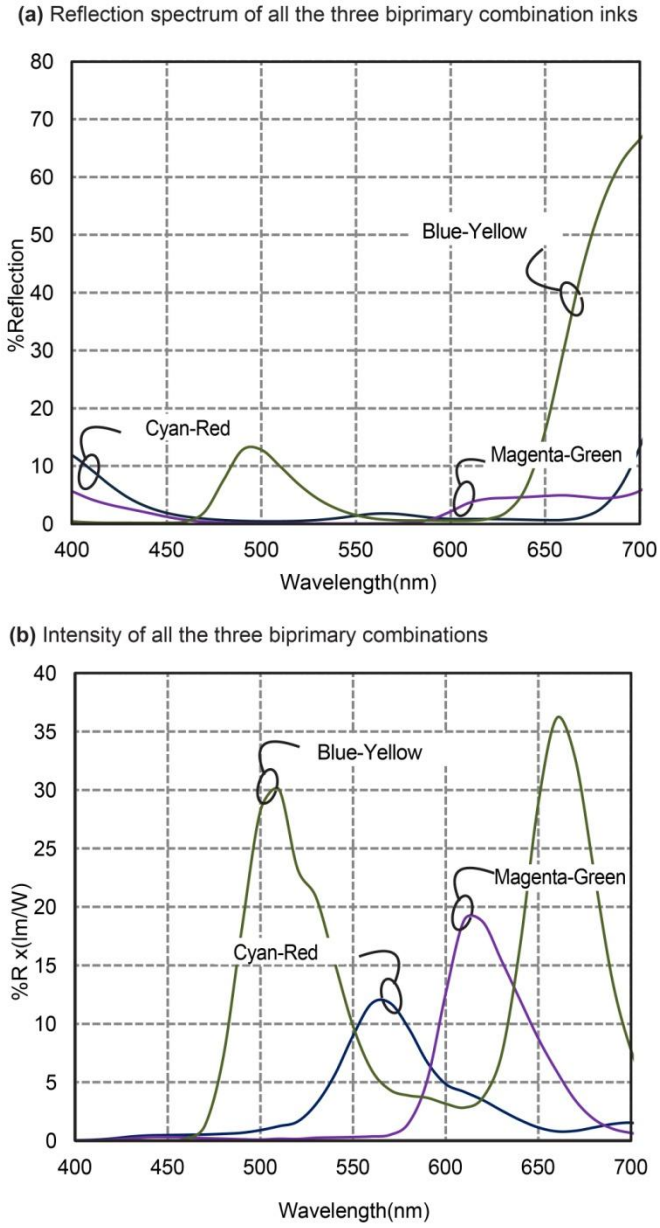



**Figure 3.9**: (a) Reflection plot of all the 3 biprimary theoretical K states (b) Intensity plot of all the 3 K states

### 3.6 Conclusion

We have successfully demonstrated here the use of a biprimary color system with G/M dual particle dispersions in both an in-plane electrophoretic pixel and in EKD pixel architecture. The results are preliminary, with the main areas of future development being creation of ink dispersions optimized for biprimary operation. Theoretical color-space analysis was also performed, and reveals the potential improvement to be realized as compared with conventional RGBW operation. The results are commercially compelling, as they are achieved with a single-layer technology capable of combining manufacturability, excellent color performance, and potential for high resolution and faster switching speeds.

The subsequent chapter now digs deeper into creating a more practical feasible device structure that can be created with simple and least fabrication steps possible, but functions similarly like the in-plane structure demonstrated here, switching the device in 4 colored states of KWXX' where X and X' are the two complementary biprimary colors. The chapter will deal with the design and fabrication of such a device along with the functioning of the biprimary inks followed by the characterization and results that explains the requirement of the applications in more detail: for displays and smart windows.

[The contents and figures of this chapter have been reproduced with permission from reference no. 11].

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# *Chapter 4: Electrokinetic pixels with biprimary inks for color displays and color-temperature-tunable smart windows*

## 4.1 Introduction to this chapter

This chapter provides an advanced implementation of the biprimary color system in applications where subtractive color is performed inside a single pixel to alter both the magnitude and color of reflection (electronic paper displays) or the optical transmission and color temperature (smart windows). A novel device structure can switch between 4 states: clear, black, either of two complementary colors from RGB and CMY sets, and also mixed states between one of these 4 states. The device structure utilizes an electrokinetic pixel structure which combines the spectral performance of in-plane electrophoretic devices with the improved switching speeds of vertical electrophoresis. The electrophoretic dispersions are dual-particle dual-colored and are controlled using two traditional planar electrokinetic electrodes on the front and back substrates, along with a 3$^{rd}$ electrode conveniently located at the perimeter of each unit cell. Demonstrated performance includes contrast ratios reaching ~ 10:1, reflectance of ~62%, and transparency of ~75%. For electronic paper displays, these results provide a pathway to double the reflective performance compared to the traditional RGBW color-filter approach. For smart windows, the technology provides not only control of shade (transmission) but also provides complete control over color-temperature. Furthermore, this 3-electrode device can be roll-to-roll fabricated without need for any alignment steps, requiring only a single micro replication step followed by self-aligned contact printing of the 3$^{rd}$ electrode.

## 4.2 Background

Light valves used in displays [1][2][3] or in smart windows [4][5] are typically limited to electronic switching between two states, typically a clear and a black state as already been stated. For

full-color operation, color filters are utilized which typically reduce the optical transmission or reflection by 3X [6]. Furthermore, even the clear to opaque switching mechanism can be inefficient itself, further limiting the optical performance to the point of preventing commercial success for applications such as color reflective displays (e-Paper)[6].

Previously we have proposed a new biprimary color system [7] in chapter 2, and we demonstrated basic proof of principle using traditional in-plane electrophoretic display architecture [8][9] in chapter 3. The biprimary color system uniquely allows a single pixel to be switched through mixtures of color states of K (black), W, (white) X and X', where X and X' states are the two complementary colors from the RGB and CMY primaries (e.g. R/C, G/M or B/Y). As demonstrated [6][7], the performance doubles both the reflectance and color saturation for reflective 'e-Paper' displays[6]. However, several significant drawbacks still existed from an applied perspective. Firstly, in-plane [9] electrophoresis is slow due to long-path lengths. Secondly, the need for at least 3 electrode controls is complex and difficult to reliably fabricate, especially when low-cost and large area devices are required [8].

This chapter report here electrokinetic pixels with biprimary inks as demonstrated in a recent paper by our research group [10], resolving several of the previous challenges in use of biprimary color. The device structure utilizes an advanced electrokinetic pixel structure which for bi-primary color-to-color switching states, combines the spectral performance of in-plane electrophoretic devices with the improved switching speeds of vertical electrophoresis[11][12][13]. The electrophoretic dispersions are dual-particle, dual-colored [14] and are controlled using two planar electrokinetic electrodes on the front and back substrates, along with a new 3$^{rd}$ electrode provided at the perimeter of each unit cell. The performance includes contrast ratios up to ~ 10:1, reflectance of ~62%, and transparency of ~75%. These results provide a pathway to double the reflective and color-fraction performance [6] compared

to traditional color-filter approaches for electronic paper displays. Because switching speeds are not fast (seconds to 10's of seconds), signage is the most likely e-Paper application [6].

A further suggestion is put forth that with single biprimary ink dispersion such as B/Y; this simple single-layer technology provides previously undemonstrated capability for smart windows. Not only can optical transmission be shaded, but uniquely, complete control over color-temperature is provided. This control over color temperature is a significant value-add for windows as a source of external light, mirroring the value already existing in color-temperature for light bulbs, and providing value that does not exist in mechanical shading. Especially important for large-area applications such as windows, the 3-electrode device can be roll-to-roll fabricated without the need for any alignment, requiring only a single micro replication step followed by self-aligned contact printing of the 3$^{rd}$ electrode. Furthermore, this approach is extendable to independent control of privacy/shade and of visible/infrared transmission.

### 4.3 Device Design, Fabrication, and Materials

Fig. 4.1 shows the top view of the devices and the cross-sectional view of two variations of the device construction, each of which function identically. The device uses electrophoresis to attract charged colored particles to the top $In_2O_3$:$SnO_2$ (ITO) transparent electrode (spread to display their color), or to compact (hide) them inside the pits adjacent to the bottom ITO transparent electrode, or compact them at the hexagonal metal electrode found along the perimeter of cell.

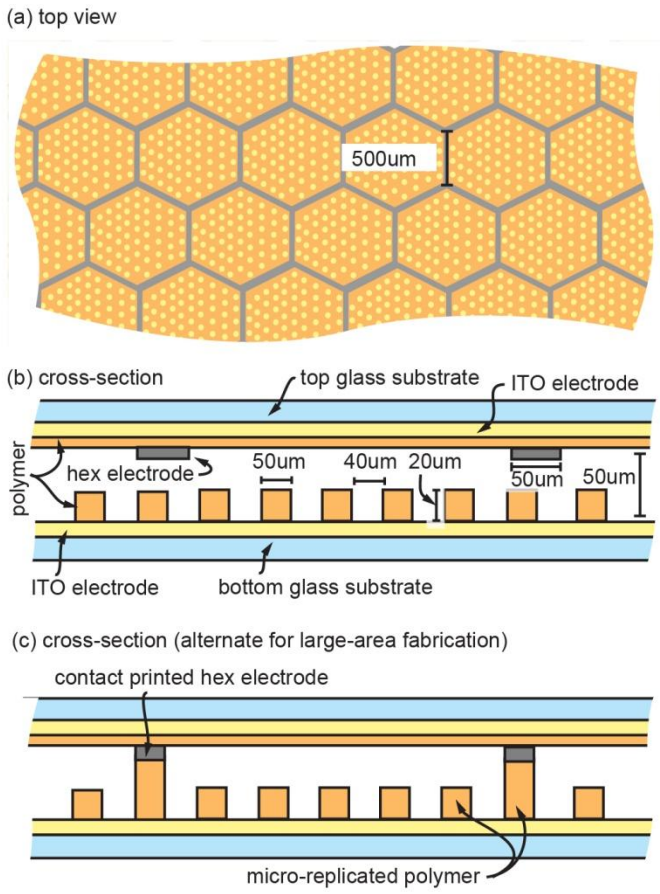


**Figure 4.1**: Diagrams of device construction with dimensions including a (a) top view and (b), (c) cross-sectional views of two different methods of fabrication where (c) can be potentially made roll-to-roll without any alignment steps. Note, the device is shown without filling of the electrophoretic ink dispersion.

The tested devices were fabricated on 2”x2” aluminosilicate glass substrates with a net active area of 1.6”x1.6”. The devices were filled with dual-color, dual-particle colloidal dispersion inks supplied by Merck Chemicals Ltd. [14], by placing a droplet of the dispersion in the center of a first substrate, and then clamping the second substrate to the first with binder clips. The particles are polydisperse, such that their electrophoretic mobilities are dissimilar but around $5x10^{-6}$ $cm^2/V\text{-}s$, as previously reported [8]. Differing mobilities lead to different velocities and therefore allow incomplete transport (spreading) of the particles if the application of voltage is

limited in time. The importance of this spreading will be presented in the next section on device operation. The raw spectral properties of the colloidal dispersion inks themselves, measured external to a device, can also be found in our previous chapter [8]. Both transmissive and reflective devices were created and tested in this work. The as-fabricated device is inherently transmissive mode. For the reflective mode, placed behind the device was a 3M ESR reflector film coated with a diffuse barium sulfate powder, resulting in a >96% Lambertian reflectance. - Because this reflector is decoupled from the device (air gap), there are no issues with light-out coupling due to total-internal reflection [15], and the expected reflectance will therefore be roughly the square of the transmittance.

There are two variants of fabrication. The first approach, shown in fig. 4.1b is useful for research purposes as the device features are formed using purely conventional photolithographic means (the polymer in Fig. 4.1b is SU-8 which is an electrically insulating negative tone photoresist). The second approach, shown in Fig. 4.1c is designed for low-cost and highly scalable manufacturing. Hewlett Packard (HP) has already shown that the lower micro replicated polymer can be made using simple roll-to-roll cast and cure technology [11-13]. The raised (higher) cell-perimeters can then be coated using simple and proven contact printing (transfer) of a conductive Ag epoxy from a roller coated with the Ag epoxy. The process used for Fig. 4.1c is highly attractive, because the entire process can be implemented roll-to-roll and requires no alignment. In addition, the Ag epoxy can also be used to provide a closed-cell seal to prevent pigment particle movement from cell to cell which is important for applications such as windows where gravity could cause a net pigment particle migration due to gravity. Further supporting the simplicity of fabrication, unlike our previous in-plane designs [8], the hexagonal electrode grid is highly fault tolerant through electrical path redundancy (open-circuit defects in the lines are a non-issue). The only meaningful differences between the design of fig. 4.1b and fig. 4.1c, is that: (1) fig. 4.1b is more likely capable of higher resolution features which are

important for displays; (2) fig. 4.1b requires additional channel height spacers (not shown); (3) the hexagonal electrode of fig. 4.1b has more exposed surface area to support compaction (greater hiding) of charged colored particles on it. The device of fig. 4.1b was utilized for the research results shown herein.

The Figure 4.2 shows the entire sequence of switching this particular device, as the switching is complicated spectrally, the most optimum switching sequence is shown:

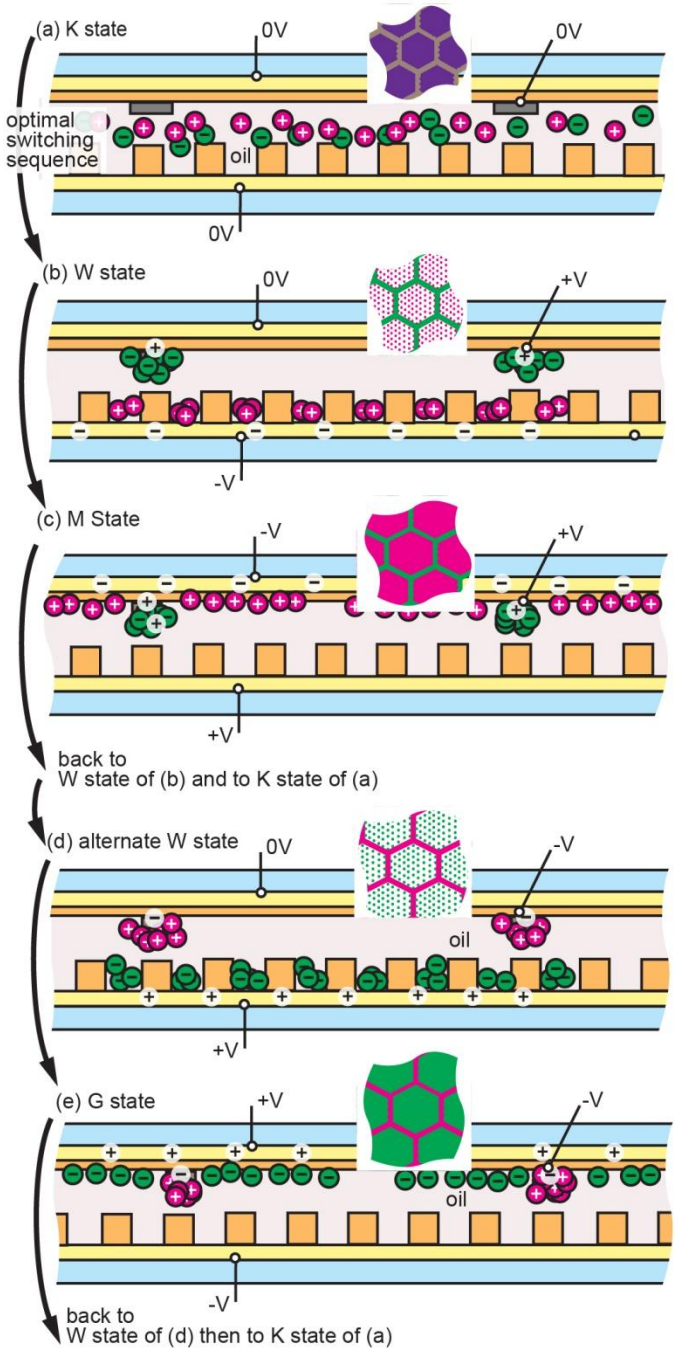


**Figure 4.2:** Device operation in (a) K (black), (b) W (white), (c) M(magenta), (d) alternate W (white), and (e) G (green) states with inset diagrams of the top view. With the current device dimensions and ink dispersions, the sequence shown was found to be optimal for switching between states.

### 4.4. General Physics of Operation

The general physics of operation are now discussed for devices, covering both reflective and transmissive modes of operation. The biprimary system *for transmissive smart-windows* uses only one color set, such as B/Y, and requires no sub-pixelation. The biprimary system *for e-Paper displays* utilizes 3 sub-pixels, each sub-pixel having one dual-color, dual-particle ink dispersion of R/C, G/M or B/Y colors [8]. In each sub-pixel, the complimentary colored particles are oppositely charged, with an example for the G/M sub pixel shown in fig. 4.2.

For the G/M example shown in fig. 4.2, the four states of KWGM are illustrated, including two possible W states. For the experiments reported here, the DC voltages applied to the electrodes are limited to only +25, -25 and 0 V. The sequence of states (KWGM) is shown in the optimal order for the fastest and most complete transitions for the current device geometry and ink dispersions, and other transitions have slower switching speeds or poorer color performance. Generally, switching that compacts a particle first into the pits, is the ideal state before the particle is then spread across the top substrate. Note, the preliminary inks used here have particles with electrophoretic mobilities that are one order of magnitude lower than the best commercial electrophoretic ink dispersions. Because switching time is proportional to electrophoretic mobility, much faster speeds are possible in future work, as is improved particle color saturation as well. A M/G example for sequential switching through various states is now described in greater detail.

***K State*:** The initial state with no voltages applied is black as the particles spread evenly over time in the absence of voltage. Actively switching to the black state will be described at the end of this section.

***W state*:** Starting from the previous K state, a W state is achieved by setting the bottom ITO electrode to -25V and the hexagonal electrodes to +25V, and the top ITO electrode is kept at

0V. As a result, negatively charged green particles are compacted at the hexagonal electrode and the magenta particles are compacted in the pits. After the switching is adequately complete (~10s) this state can be maintained with voltages of less than 10V.

***M state*:** Next a M state is achieved from the previous W state by switching the bottom ITO electrode to +25V and the top ITO electrode to -25V which then draws the M particles to a spread state on the top plate, while the hexagonal electrode is still maintained at the same +25V from previous state, retaining compaction of the G particles. The switching is complete as perceived by the naked eye at a distance, in (~10s) and can be maintained at a low holding voltage of +/-10V.

***Alternate W state*:** The alternate W state is where the negatively charged green particles are compacted in the micropits by setting the bottom electrode to +25V and compacting the M particles to the hexagonal electrode by setting it to -25V while keeping the top ITO plate at 0V. As shown in Fig. 2, for optimal optical performance (most complete particle movement), it is best to leave the M state (2c), return to the original W state (2b), then actively to the K state (see section below), then finally switch to the alternate W state. Again, this state can be maintained at a low holding voltage of +/-10V.

***G state*:** Next the G state is set from the previous W state by reversing the bottom ITO plate to -25V while the top plate is switched to +25 V which moves and spreads G particles to the top. The hexagonal electrode is kept at the same potential of -25V from the previous state which still holds the M particles in their place. This state can then be maintained at a low holding voltage of +/-10V after the switching is complete in ~10s.

***Actively Returning to the K state*:** A multi step process is needed similar that needed for the alternate W state, for optimal optical performance (most complete particle movement). It is best to first leave the G state (2e) by returning to the alternate W state (d). Then the K state can be

achieved within ~7 s by spreading the particles through repeatedly reversing voltage between the bottom and the hex electrodes (Fig. 4.1), followed by removing all the voltages. Complete mixing is difficult to quantify and is achieved only after the voltages are removed for 10's.

## 4.5 Experimental Results

### 4.5.1 Reflective results (e-Paper applications).

Reflective measurements were made utilizing an Ocean Optics HR4000CG-UV-NIR spectrometer. The sample device is illuminated with white light using THORLAB OSL fiber illuminator. The spectrum measured is specular-excluded, and collected by an Ocean Optics P200-2-VIS-NIR fiber fitted with a 74-VIS collimating lens. The set-up is calibrated against Labsphere Spectralon reflection standards of 99% and 2% for bright and dark spectrum, respectively. The reflection spectra of all the KWGM states were measured in the device and plotted in Fig. 4.3 along with zoom-in photos of the states. The best achievable W state was found to be ~62% reflective with a K state of ~6%. The G and M states are much more saturated than that was demonstrated for the in-plane structure [8] due to pigment compaction at multiple pits, and due to shorter path lengths in moving to the spread state. The colloidal ink dispersions have not been optimized for this particular device structure and therefore with such optimization, even better performance is fully expected for color saturation, black state and white state. Even so, right now, a reflectance of ~62% with biprimary color already provides more than twice the color performance [6] compared to RGBW color filters on e-Ink technology (vertical electrophoresis, ~20% reflectance for white)[6].

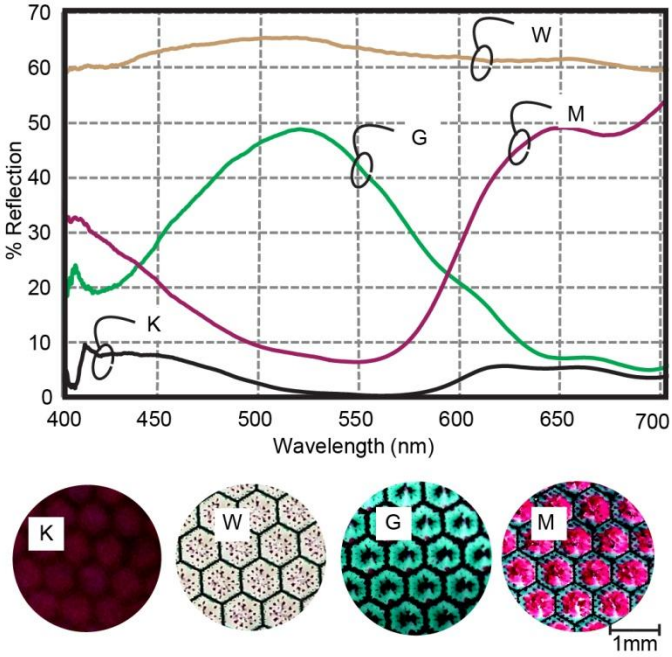


**Figure 4.3**: Reflection spectra of a device with G/M (green/magenta) particle dispersion, along with inset photographs of each state. Data was obtained by placing a 96% diffuse reflector beneath the device structure shown in Fig. 4.2.

### 4.5.2 Transmissive results (smart window applications)

For transmissive operation, an R/C colloidal dispersion ink was chosen as pictured in Fig. 4.4, because the other ink dispersions (M/G, Y/B) currently lack the opacity needed for a good black transmissive state.

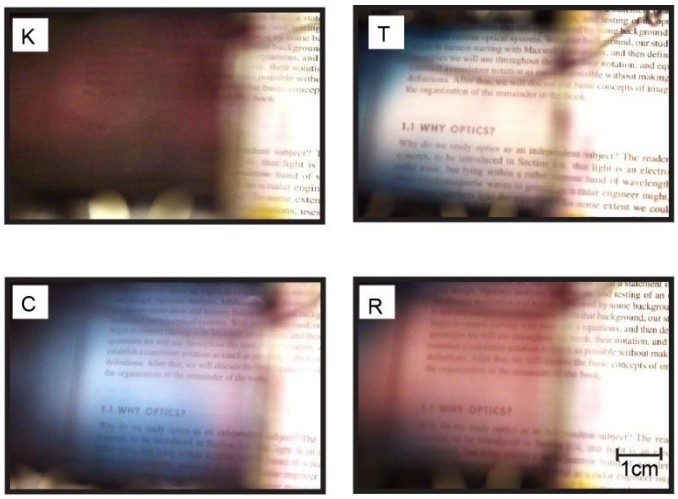


**Figure 4.4**: Zoom-out photographs showing all the KTCR (black/transmissive/cyan/red) states for the RC (red/cyan) dispersion in a transmissive mode device. An optics textbook is provided behind the device to validate clarity of transmission and transmission efficiency

Like the reflective results, optimizing the ink dispersions and/or constructing a thicker channel should both improve the opacity of the black states. The dispersions and device were originally developed for reflective operation, which has twice the optical path length of transmissive mode. Of particular interest for windows, is improving the Y/B ink dispersions for transmissive color-temperature control.

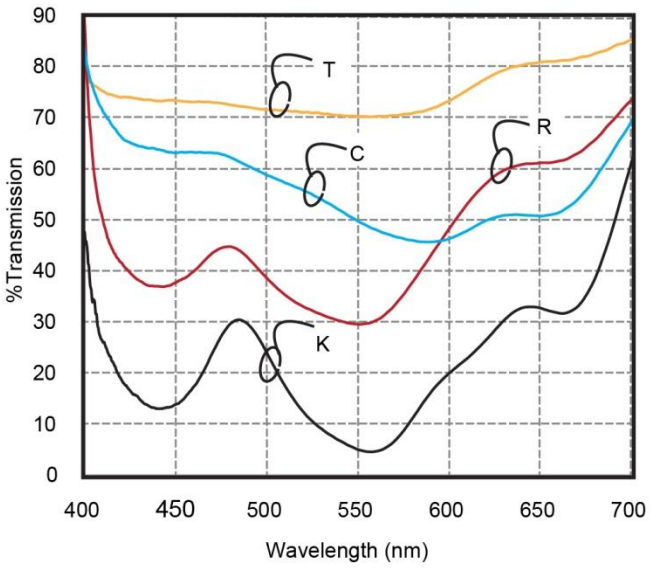


**Figure 4.5:** Transmission spectra of the R/C (red/cyan) dispersion measured in the device for all the KTCR states (black/transmissive/cyan/red)

Transmission measurements were also made utilizing the Ocean Optics HR4000CG-UV-NIR spectrometer and a fiber illuminator. The incoming transmitted spectrum through the device is collected by an Ocean Optics P200-2-VIS-NIR attached to a 74-VIS collimating lens. The set-up is calibrated with respect to air, hence the transmission spectra includes the light absorption and Fresnel reflection due to the stack of the device (~12% of the incoming light). The transmission spectra are shown in Fig. 4.5 for the 4 states of K (dimmed) T (clear white), C (cyan) and R (red). The clear white transparent (T) state is found to be ~75%, comparable or better than the transparency of other technologies that exists for the smart windows [16] [17].

### 4.5.3 Experimental and Modeling Results for Pigment Compaction

To further understand the device operating principles, additional modeling analysis was also performed. One question of particular interest was to determine how charged particles could transition from compaction on the hex borders, to an adequately even spreading across the cell or into the micropits across the cell. The optimum switching sequence shown in fig. 4.2 was selected because of this very issue, which is now presented here in more detail through supplemental modeling work. A COMSOL™ model was used to investigate unit cells as shown in the Fig. 4.6(a). The 2D simulation pictures (b)-(e) show the E-field lines (arrows) and the electric potential (color gradient) and therefore shows the path through which positive particles will move. Conversely the negative particles will move in the reverse direction toward the arrowheads. The arrows above the hexagonal electrode can be ignored because in the device a solid dielectric is located there.

The simulation result of W state of fig. 4.2b is shown in fig. 4.6b. The behavior of particles moving from the K state of fig. 4.2a to the W state of fig. 4.2b is conventional and the simulation simply confirms the expected and observed behavior. For switching from the W state of fig. 4.2b to the M state of fig. 4.2c, the simulation shown in fig. 4.6c is a simple result and expected.

Now, the transition from the M state of fig. 4.2c to the alternate W state of fig. 4.2d is not conventional. For switching it from M to alternate W state the M pigment needs to move in-plane which is slow, but conventional. However the G particles which were already on the hex electrodes will move straight down to the nearest pits if one attempts to go straight from the M state to the alternate W state. This can be seen also as a path of strong E-field as shown in fig. 4.6(d). Hence, intermediate states of switching, as shown in fig. 4.2 are necessary to properly spread the G particles and complete the transition to the alternate W state.

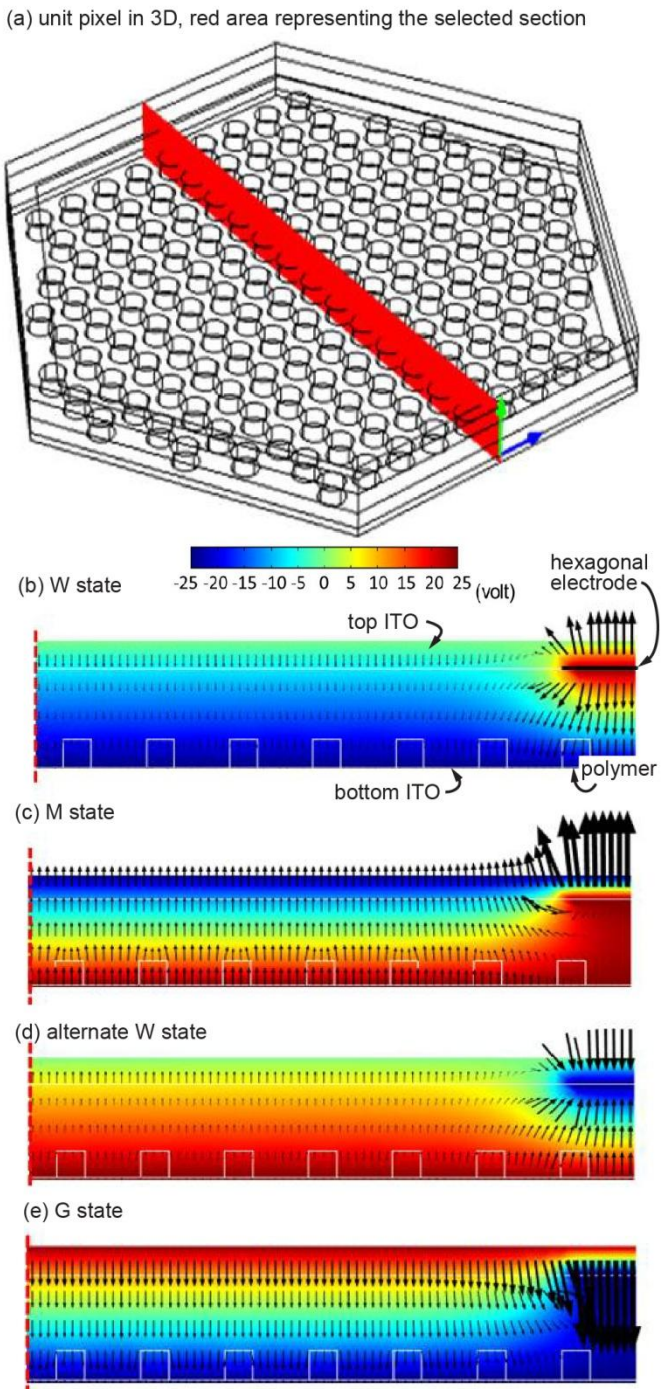


**Figure 4.6**: (a) 3D model of a unit pixel (b)–(e). Simulation results of the states showing the E-field path for W, M, alternate W, and G states.

Next, from the alternate W state to the G state, the G particles are spread and the switching is conventional and simple/expected (fig. 4.6e). Lastly, from the G state to the K state, there are similar challenges to that of switching to the alternate W state (fig 4.6d) and a multi-step switching process is required.

The simulation and above discussion highlights the need for a more optimal arrangement of the micropits. The micro pit array is evenly spaced, as designed for conventional single-particle electrokinetic display operation. A higher density of the micropits, or larger micropits, towards the perimeter of the pixel would be expected to provide a faster switch to high optical transmission, as the pigment will have a shorter path to travel for compaction after leaving the hex electrode. This and other modifications are possible, such as also optimizing the hexagonal electrode pitch relative to the micropits arrangement, but are beyond the scope of this work. Other possible improvements include use of index-matched ITO and index-matching of the fluids to the surrounding layers and particles (less Fresnel reflection, less scattering), and the significant need for optimization of the particle dispersions themselves.

### 4.6. Brief Discussion and Mock-up for Smart Windows

Application for smart windows is discussed in greater detail, an application where the combination of simple fabrication and increased optical performance is likely most compelling. The most important colloidal dispersion ink- for smart windows would be B/Y. Smart windows based on Electrochromism [16] or polymer dispersed liquid crystals [17] have struggled to find broad-based commercial success partly because their performance value over traditional mechanical blinds is not significant. Windows are utilized as a form of lighting, and the commercial market has been sensitized for decades to demand lighting with a variety of color temperatures (bluish vs. yellowish light). Therefore, the devices demonstrated here, along with an improved B/Y dispersion, could provide the first ever smart window capable of both dimming

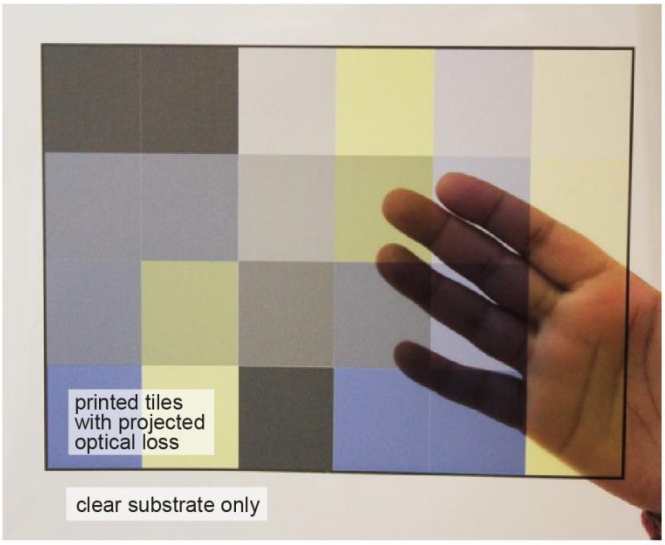


**Figure 4.7**: Blue/yellow printed mock-up of the window application, the colors showing blue, yellow, gray and all the intermediate shades. The mock-up includes the expected optical loss due to pixel borders and micropits.

light transmission, but also fully altering the color temperature of transmitted light. The yellow-state could even be amber/orange tinted, which would likely only improve the opacity of the black state.

Fig. 4.7 shows a printed mock-up of this technology, including the optical losses expected by use of conventional pigment particles, due to factoring in loss expected for the area of the micro-pits and hexagonal electrode. The mock up shows a variety of clear, black, gray, and color-temperature modified states. More detailed spectral measurements of actual blue and yellow particle dispersions can be found in the previous chapter [8].

Lastly, it should also be noted that with other types of two particle dispersions, windows could potentially be created with independent control of shade and privacy (black particles, and white scattering particles) or independent thermal control (black particle, clear but IR absorbing particle). In fact, high performance two-particle dispersions such as black/white are easier to

create because the particles are chemically very distinct from each other on their surface, and are readily available commercially (e.g. used in E-Ink technology). These black/white particle dispersions have ~10X higher electrophoretic mobility value than the dispersions used in this work along with excellent long-life stability.

### 4.7. Conclusion

We have successfully demonstrated in this chapter a new 3-electrode electrokinetic device structure with dual-particle, dual-color colloidal dispersion inks for reflective E-paper displays and smart windows. This device adds the ability of switching pixels into multiple spectral-states, potentially improving color performance in reflective displays, and providing color temperature control for smart windows. The switching speeds are on the order of 10s or more, likely relegating e-Paper use to signage applications. For smart windows, these speeds are similar to those commercially found in electrochromic windows. The results shown herein already show a significant boost in transmission, reflection, and color performance, despite the fact that the dual-particle, dual-color dispersions are far from optimized performance, nor has the device structure been optimized. In addition, the devices involve very few and simple fabrication steps, such that low-cost and large area fabrication is quite feasible. Of all the device aspects, particle dispersion improvements are likely most critical, especially in the areas of spectral properties, switching speed, and particle/dispersion stability.

This work completes the overall aim of this dissertation, by providing a novel approach that in optical performance clearly advances the state of the art in large-area light valves, and importantly does so with very simple and inexpensive fabrication techniques.

[The contents and figures of this chapter have been reproduced with permission from reference 10.]

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# *Chapter 5: Future work and improvements*

## 5.1 Introduction to the chapter

The previous chapters discussed in detail how we can create devices using electrophoretic movement of dual-color particles (biprimary color) to switch the pixels in multi-spectral states (K, W, X and X'). These chapters cover the fundamentals of how a single layer biprimary color system can be used to implement multi-switching of pixels catering to the development of the full color reflective displays and color tunable transmissive smart windows. However, some future work and improvements are suggested in this chapter to enable this technology to achieve commercial use in displays or windows.

## 5.2 Reflective Display applications and improvements

### 5.2.1 E-readers

We already discussed e-readers in chapter 1 and how currently the commercial product that dominates the market is the Amazon Kindle™ that uses E-ink. The black and white displays being popular among users, there is an increasing demand for color versions of the same display. However, E-ink as already discussed has not yet provided any compelling way of generating a color version of it, and we explained how the biprimary color system if used for a technology like this will be able to provide full color operation in a single layer system [1]. Our technology as discussed in this dissertation has been shown to be able to switch one pixel into 4 color states. Although the switching time is slow, this technology can still be used for products that do not need a video operation, such as an e-reader or signage. With improved and optimized electrophoretic mobility values which will be higher than the current electrophoretic mobility of the dispersions, a better switching time of about 1 second is likely to be achieved.

**Figure 5.1**: The future of e-paper devices where a user unfolds a flexible mobile device to a large paper format color display to watch a video or animation.

This may not be fast enough for e-readers, but is more than adequate for signage applications. Longer term, if the device dimensions can be reduced, switching speeds could reach several hundred ms, allowing e-Paper products to be created for the first time, such as that depicted in Figure 5.1.

### 5.2.2 Electronic shelf labels

Electronic shelf labels are increasingly being used in displaying pricing of items in supermarkets and stores. This enables dynamic control of updating the pricing of products as well as allows the retailers to better respond to the market pricing. The requirement of using a display technology that will suffice includes both long battery life and cost effectiveness for which the retailers need to make the investment into thousands of tags in the store. The most commonly used display type for this application is reflective LCD due to the fact that it can be very low cost. Several new devices are also now commercial. First there is a new E-ink 3 particle displays (black/white and red) which uses vertical electrophoretic switching. There are also now ZBD displays using reflective LCDs that are also bistable [1]. These newer shelf-label

technologies are shown in Figure 5.2. It should be noted, that although the E-Ink device switches 3 particles, it cannot achieve robust biprimary operation because mixed states are challenging to implement.

The proposed and demonstrated biprimary technology of this dissertation can likely be a higher performance replacement for existing ESL technology. Also the slower switching of this technology will not likely be an issue since the ESLs are updated only daily or even less. But improvements will be needed for the driving schemes required to drive these displays, as will be discussed later in section 5.4.

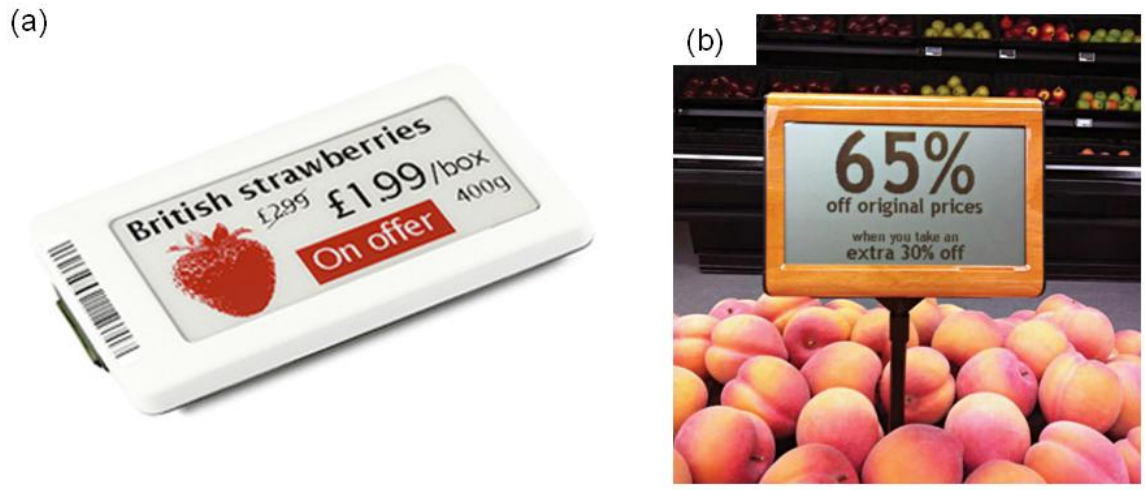


**Figure 5.2:** (a) The new E-ink 3 particle displays for shelf labels and (b) bistable ZBD ESLs in a supermarket

### 5.2.3 Bill-board signage

Large area bill-board signage that is slow in switching speed is another important color e-paper application. The biprimary color system with this device structure demonstrated in this dissertation will likely be a compelling technology for this application due to its color output in reflective mode [2][3][4]. The billboard signage does not require fast switching time but needs good color output, especially for advertising applications. Figure 5.3 shows a larger area sign by E-ink. Cholesteric LC billboards are the only technology used at true bill-board type dimensions, but that product did not succeed commercially due to inadequate color performance. Clearly there is an opportunity for biprimary color in billboards. The biggest challenge here is large area manufacturing. Importantly, a central advantage as presented in Chapter 4 is the ease of fabrication of devices using roll-to-roll fabrication with no-alignment. However, for full color operation, alignment and sub-pixelation are needed which complicates fabrication significantly. Therefore full-color billboard type signage might not be a strong application for the results of this dissertation.


**Figure 5.3:** A large area signage by E-ink

## 5.3 Transmissive applications and improvements

### 5.3.1 Color temperature tunable smart windows

The independent control of color temperature of light along with a dimmed and a transparent state is a new concept which has not been previously demonstrated [3]. This adds a much more compelling functionality compared to already existing technologies for smart windows in the market. The detail of this application has already been explained in the previous chapter. Mockup images of what the performance could look like is shown in Fig. 5.4. Subsequent improvements of the particle dispersion and optimized performance in an electrokinetic structure will improve the functionality even more.


**Figure 5.4**: The color temperature tunable windows demonstrated for (a) neutral (b) tinted (c) cool and (d) warm windows [picture credit: UC and Tim Zarki]

One factor that could be important when using this technology for window applications is the effect of ultra violet spectrum that is coming from the incoming solar spectrum. Ultra violet light can degrade organic/polymeric type materials over time, and windows often are expected to last for decades before being replaced. This will be another research parameter to consider while optimizing the particle dispersions and their stability under the effect of light spectrum beyond the visible range. One approach that would be helpful would be to apply a UV absorbing layer to the external surface of such a window.

### 5.3.2. Complementary spectrum tunable smart windows (energy control)

Most smart glass technologies like electrochromic [5] or PDLCs [6] alter incoming solar spectrum to either opaque or clear. One application that cannot be done by these prior device structures is a smart window that can alter transmission between the visible and infra-red spectrum of solar spectrum. For this application the particle dispersion required need not be biprimary but such that one of the particle types blocks *only* the visible range spectrum (e.g. using a pigment like Parylene Black) and the other particle type only blocks the infra-red (heat) spectrum (infrared pigments or dyes embedded in pigments) as depicted in Figure 5.5. This application will be particularly important in areas where temperature changes rapidly between seasons. One can choose to have control over letting the light spectrum but also block the heat spectrum or vice versa, or both or none. Therefore this application is possible adding to a functionality that will be useful in both energy savings as well as privacy control.

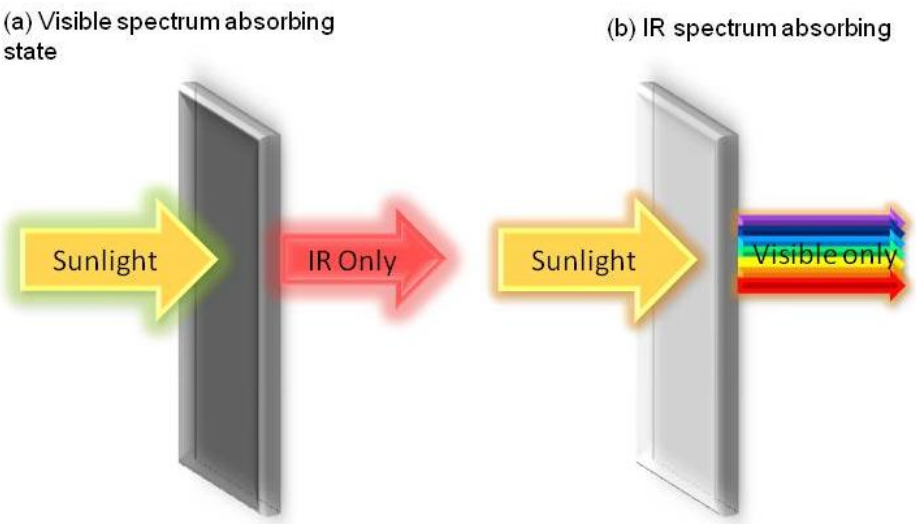


**Figure 5.5**: An energy controlling smart window (a) visible absorbing and (b) IR absorbing

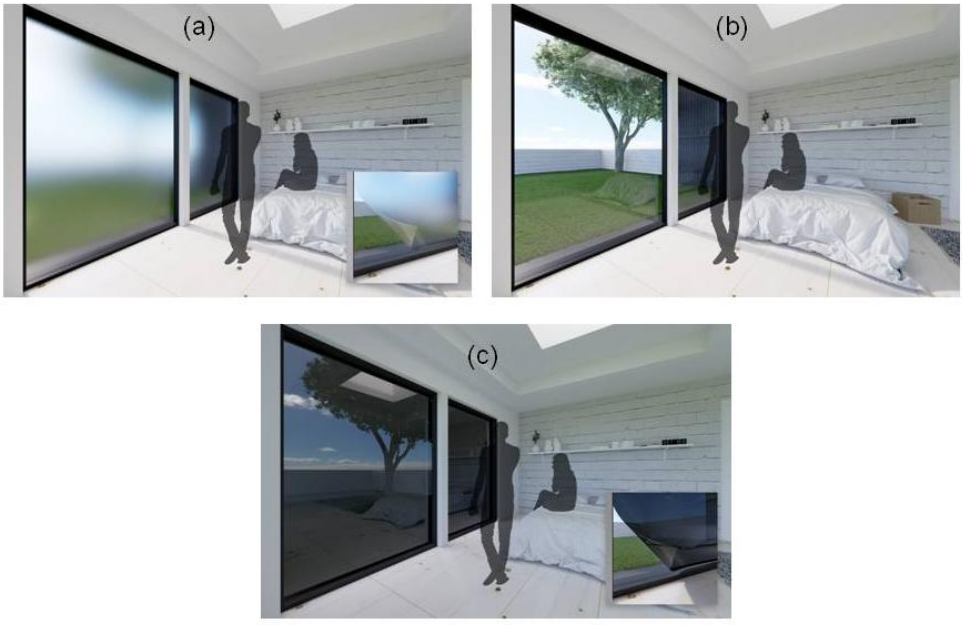


**Figure 5.6**: A privacy controlling smart window showing privacy control in (a) frosted shading (b) no shading and (c) tinted shading [picture credit: UC and Tim Zarki]

### 5.3.3 Privacy controlling smart windows

Smart windows like the polymer dispersed liquid crystals (PDLCs) [6] switches between hazy and optically clear states and are popular for privacy control. This application can be even better served by the technology that we have demonstrated in this dissertation. By utilizing two different types of particles, such that one particle is optically scattering (particles with a refractive index ~2.0) and the other is opaque (black), then different shades of privacy and dimming can be utilized for home windows in place of window blinds for privacy (Figure 5.6). This provides added advantage that you can have privacy, but let all the light in, or you can dim the light as desired. There is no technology that can currently enable this.

## 5.4 Driving schemes

To electronically drive the technology of this dissertation, it is important to explore the driving schemes that are normally employed to drive display pixels. We review some of the basic driving schemes that are normally used in displays, and also discuss which of these might apply to windows as well.

### 5.4.1 Direct drive

The simplest and the most applicable and easily implementable driving scheme is the direct driving for all display types, by applying voltage directly to a number of electrodes by laying out a fan of electrodes for each pixel. For all the experiments conducted in this dissertation, the kind of driving used was the direct driving, where each electrode was driven individually (Figure 5.7). This is an old method of driving displays and newer generation commercial displays do not utilize this method of driving. For the smart windows demonstrated in this work there is no need to use sub pixels and a single biprimary dispersion pair can be used to switch the window in 4 color states. Therefore, direct driving may be continued to be useful for windows, but not for display applications.

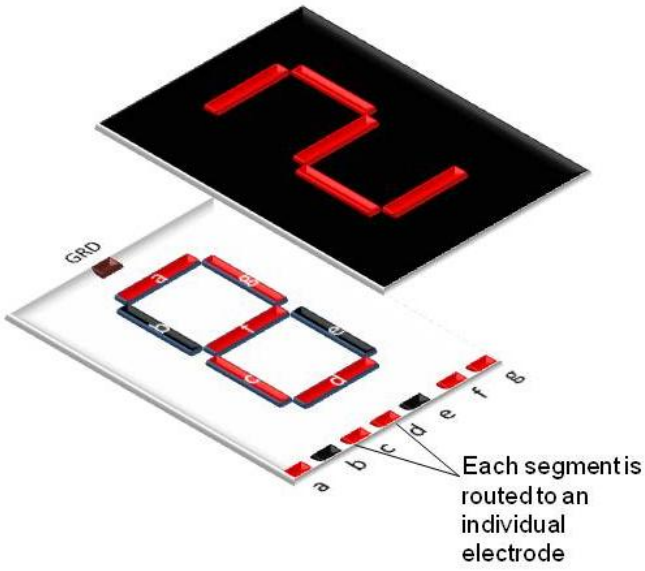


**Figure 5.7:** A seven segment display showing direct driving (each segment routes to individual electrodes and driven directly)

### 5.4.2 Passive matrix

Passive matrix driving can achieve medium resolution displays and are implemented because they have a low cost fabrication process. The electrodes are laid out in rows and columns orthogonally to each other. A potential is applied to activate a specific pixel at the intersection of these orthogonal electrodes (Figure 5.8). The technology described for reflective displays will have 3 biprimary sub-pixels, and to drive that with a passive matrix scheme would require deeper research for an effective driving/holding scheme. Optimization of the device designs with pulse times and voltage applied will need more investigation. In addition, the inks may require some bistability to help hold the image while the columns and rows are scanned, and when particles are to be spread uniform AC voltages would be applied above the threshold for particle movement.

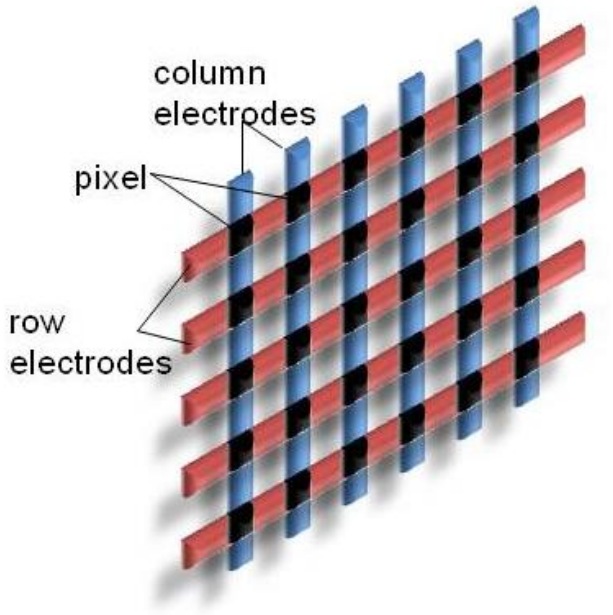


**Figure 5.8:** A typical passive matrix display arrangement with row and column electrodes

### 5.4.3 Active matrix

Active matrix driving is the most common driving method to drive displays nowadays. This method provides one of the highest resolutions. They generally constitute a backplane of thin-film transistors (TFT) connected to the individual pixels and can be individually addressable (Figure 5.9). The only possible drawback with this driving scheme is that they need to be fabricated and integrated onto a TFT backplane and the operating voltage to drive them is usually between 15-20V. Higher voltage can damage the TFTs. Fortunately, the devices of this dissertation are compatible with 15-20V operation, even more so with improvement of the particle dispersions (e.g. higher electrophoretic mobilities).

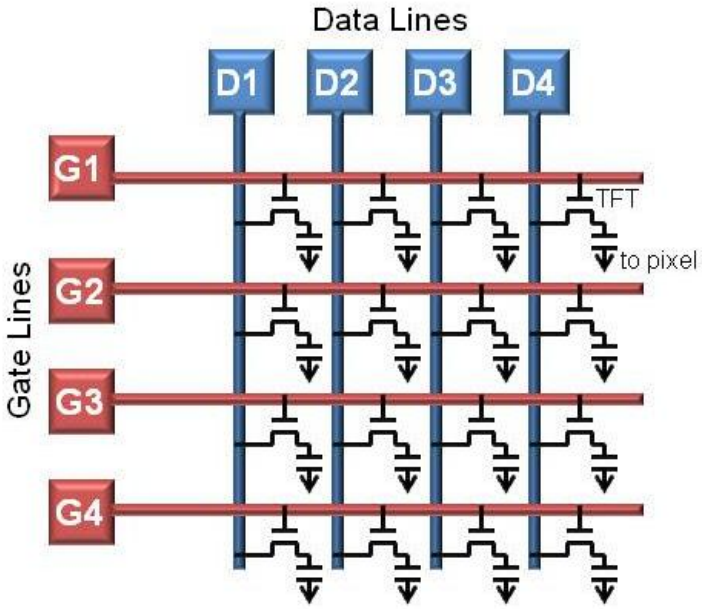


**Figure 5.9:** An Active matrix display (4X4) driver showing the data lines and gate lines connected to the TFTs.

For E-inks electrophoretic displays they have used active matrix driving, and HP's electrokinetic structure that constitutes two planar front and back electrode could also be driven by active matrix. So the device structure described in this work will need investigation when employing active matrix driving for their operating voltage and connecting the TFT backplanes (3 electrodes instead of 2).

### 5.5 Device sealing and Roll-to-Roll processing

One more issue that needs consideration is the sealing of the device assembly. Most of the experiments were conducted with unsealed devices that are held by applied pressure (binder clips). This was possible as the device need not be aligned by its top and bottom plate. But the process of assembling and sealing this device with the particles inside for commercial applications need to be considered. One such process is described here for flexible substrate fabrication and sealing for the device described in the dissertation.

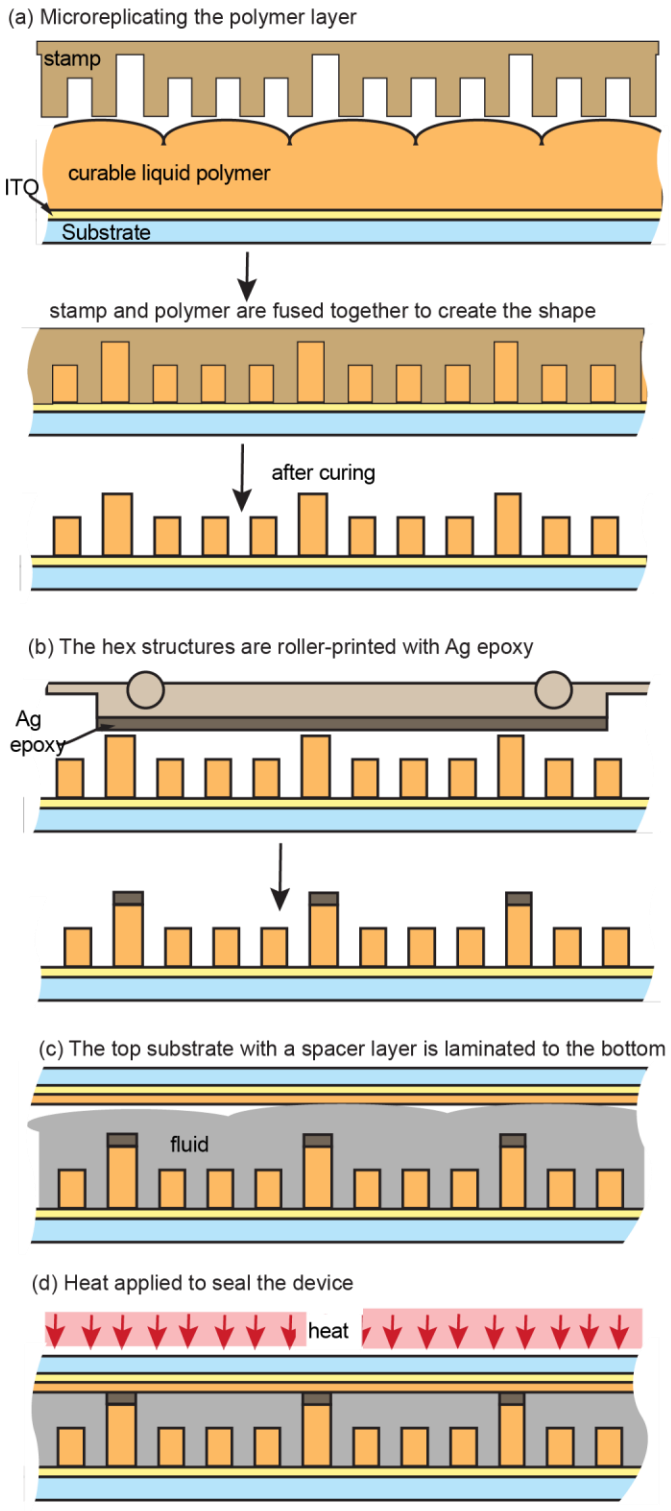


**Figure 5.10**: A process diagram showing step-by-step method of fabrication of the device on a flexible substrate using micro replication and printing only.

The bottom polymer layer is micro-replicated for the micropits structures as shown in the figure 5.10. The polymer layer that is micro replicated is cured. Next the hexagonal electrode is now roller printed as a conductive epoxy (thermal curing). The top substrate is now applied (with a spacer layer of polymer) by lamination after dosing the fluid. As the top substrate is applied the roller is hot enough to cure the epoxy and seal each cell. The electrode contacts are made by de-laminating the edge, the hex grid is contacted by using metal tape. The lower ITO electrodes can be contacted by scraping off the polymer layer.

**5.6 Conclusion**

This dissertation demonstrates the fundamentals of large area multi-spectral state electrophoretic panels that can be used for a variety of applications in reflective and transmissive mode panels. The key advantages that have been established over the current dominant technologies are using a color system called the biprimary color system that provides better color output than other single-layer technologies. This technology was improved from an in-plane 3 electrode system to a hybrid electrokinetic 3 electrode system. This development enhanced not only the color saturation and the switching speed but also simplified the fabrication to a large extent. This type of achievement is normally mutually exclusive (e.g. improved performance traditionally requires more complex fabrication). Furthermore this structure is simple enough to be able to fabricate on flexible substrates that lays the platform for foldable e-paper displays in the future. This technology is also found to be compelling for smart window application and addresses a market where windows could be essentially utilized for control of color temperature from window light, privacy and dimming control, or spectral energy (heat) control.

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# *Appendix: Fabrication process sheets*

## A.1 Fabrication of 3 interdigitated electrode structures for in-plane electrophoretic device

1. Fabrication of electrode

- A 2X2 square ITO slide is taken, and a photo resist (SU8 2002) is coated at a speed of 3000 rpm for 30 sec.
- Soft baked at 100°C for 4 min.
- Exposed for 70 sec.
- Post exposure baked for 6 min at 100°C.
- Developed in PGMEA for 20 sec. (The electrode thickness is obtained as 8.4 um).
- Hard baked for 30 min at 180°C
- Etched in a solution of acids HCl:H2O:$HNO_3$ (18% HCl, 70% $HNO_3$) 4:2:1 ratio.

1) Fabrication of spacer:

- A Layer of SU8 3020 (20 um) is coated at a speed of 3000 rpm for 40 sec.
- Soft baked for 15 min at 100°C
- Exposed for 60 sec.
- Post Exposure baked for 5 min.
- Developed for 30 sec in PGMEA.
- Hard baked for 30 min at 180°C.

2) Dielectric Layer:

- A 2 um Layer of Parylene C is deposited by Chemical Vapor deposition method as the dielectric layer.

**A.2 Fabrication of 3 electrode electrokinetic structure device:**

1. Fabrication of the bottom plate

- Glass plate of 2X2 squares containing ITO is first spin coated with SU8 20um thick at 3000rpm speed.
- The spin coated wafer is then soft baked for 6 min at 100°C on hot plate.
- The soft baked wafer is then put on the chrome mask containing the desired pattern of ITO micropits and exposed for 50 sec to UV exposer.
- The wafer is then baked for another 6 min on a hot plate @ 100 °C
- The wafer is then developed in PGMEA for 1 min and hard baked at 180 °C for 30 min.

2. Fabrication of the bottom plate:

- The top plate containing a conformal ITO is first coated with a 20um thick SU8 layer spin coated at 3000 rpm speed
- The plate is soft-baked for 6 min at 100°C
- The soft baked plate is exposed to UV for 50 sec to crosslink the resist.
- The plate is developed in PGMEA to form a uniform thick coating.
- The wafer is then spun with a layer of S1818 at 4000 rpm
- The plate is then soft baked for 2 min at 100°C
- UV exposed for 40 sec.

- Developed in Shipley developer the pattern developed is the border of the hexagonal structure.
- This plate is then sputtered coated with aluminum of a thickness of 60nm, then solvent etched with acetone in an ultrasound sonicator to form the pattern of aluminum layer.

2. The top and bottom plates are then aligned and stacked such that the hexagonal structures looks like aluminum borders with ITO micropits.